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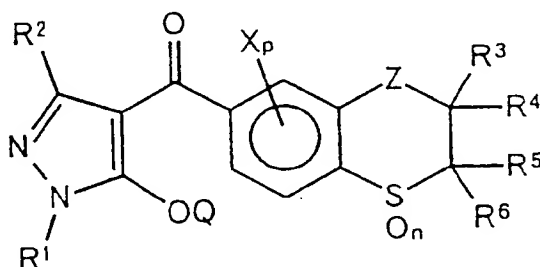
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(54) HERBICIDAL COMPOSITION

(57) Provided by the present invention is a herbicide composition containing, as active ingredients, a pyrazole derivative of the general formula (I)



(I)

Q2 X2
2nd fls, p. 38-45

and at least one herbicide compound selected from the group consisting of chloroacetamide-containing herbicides, imidazoline-containing herbicides, atrazine, cyanazine, metribuzin, linuron, metbenzuron, bentazone, dicamba, chlopyralid, 2,4-D, bromoxynil, pendimethalin, nicosulfuron, rimsulfuron, primisulfuron and pyridate. The herbicide composition of the present invention can control a broad range of gramineous weeds and broad-leaved weeds at a very low dosage without damaging corn and other crops.

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Description

TECHNICAL FIELD

The present invention relates to a herbicide composition containing pyrazole derivative.

TECHNICAL BACKGROUND

Herbicides are very important chemicals for saving weed-controlling labors and improving the yield of agricultural and horticultural crops and have been therefore aggressively studied and developed for many years, and a diversity of herbicides have now been put to practical use. However, it cannot be said that herbicides which have been so far developed have broad herbicidal spectra, and at present there are an increasing number of weeds which are difficult to control. Herbicides having broad herbicidal spectra are therefore desired. For overcoming the environmental pollution problem of conventional herbicides, further, there are desired herbicides which are effective at a further decreased dosage.

The present inventors already found that a variety of pyrazole derivatives in which a thiochroman ring is bonded to a pyrazole ring through a carbonyl group is free from damaging gramineous upland crops but can control gramineous weeds and broad-leaved weeds together at a low dosage by any one of soil treatment and foliar treatment, and filed patent applications directed to these pyrazole derivatives and herbicides containing these as active ingredients. Of these patent applications, some have been already laid-open as International Laid-open Patent Publications Nos. WO95/04054, WO93/18031, WO94/01431 and WO95/13275. Japanese Patent Applications Nos. 6-237981, 7-80059 and 7-158842 have not yet been laid-open.

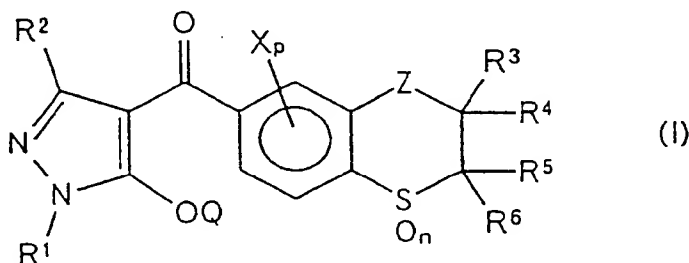
Those pyrazole derivatives disclosed in the above International Laid-open Patent Publications and Japanese Patent Applications can control gramineous weeds and broad-leaved weeds together at a low dosage by any one of soil treatment and foliar treatment, while there is desired a herbicide which can control a broad range of gramineous weeds and broad-leaved weeds at a further decreased dosage.

It is therefore an object of the present invention to provide herbicide compositions which can control a broad range of gramineous weeds and broad-leaved weeds at a very low dosage without damaging crops such as corn, etc., by using the pyrazole derivatives disclosed in the International Laid-open Patent Publications and the above Japanese Patent Applications in combination with other herbicide compounds.

The present inventors have made diligent studies to achieve the above object, and as a result have found that when a pyrazole derivative of the following general formula (I) described in the above International Laid-open Patent Publications and Japanese Patent Applications (to be sometimes referred to as "pyrazole derivative (I)" hereinafter) and a specific herbicide compound are used in combination, a broad range of gramineous weeds and broad-leaved weeds can be controlled at a very low dosage by synergistic effects of the pyrazole derivative (I) and the said specific herbicide compound without damaging crops such as corn, etc. The present invention has been accordingly completed.

DISCLOSURE OF THE INVENTION

The gist of the present invention is a herbicide composition containing, as active ingredients, a pyrazole derivative of the general formula (I),



{wherein:

R¹ is a C₁~C₄ alkyl group, a C₂~C₄ alkenyl group or a C₂~C₄ haloalkenyl group,

R² is a hydrogen atom, a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group or a C₂~C₄ alkoxyalkyl group,

X is a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group, a C₂~C₄ alkoxyalkyl group, a halogen atom, a C₁~C₄ alkoxy

group or a C₁~C₄ haloalkoxy group,

p is an integer of 0, 1 or 2,

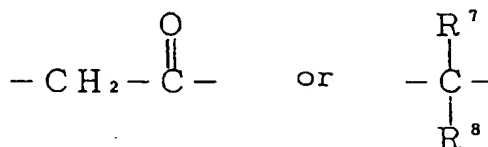
each of R³, R⁴, R⁵ and R⁶ is independently a hydrogen atom, a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group or a C₂~C₄ alkoxyalkyl group,

n is an integer of 0, 1 or 2,

Q is a hydrogen atom or a group of A-B,

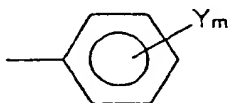
[in which

A is a group of



(in which each of R⁷ and R⁸ is independently a hydrogen atom or a C₁~C₄ alkyl group), and

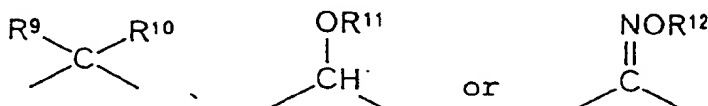
B is a C₁~C₁₂ alkyl group, a C₃~C₁₀ cycloalkyl group or a group of



(in which Y is a C₁~C₄ alkyl group, a C₁~C₄ alkoxy group, a C₁~C₄ haloalkyl group, a nitro group or a halogen atom, and

m is an integer of 0 or 1 - 3]], and

Z is



[in which

R⁹ is a hydrogen atom, a C₁~C₄ alkyl group or a C₁~C₄ haloalkyl group,

R^{10} is a hydrogen atom, a $C_1\sim C_4$ alkyl group, a $C_2\sim C_4$ alkenyl group or a $C_2\sim C_4$ alkynyl group.

R^{11} is a $C_1\sim C_4$ alkyl group, a $C_1\sim C_4$ haloalkyl group, a $C_3\sim C_6$ cycloalkyl group, a $C_3\sim C_6$ alkenylalkyl group, a $C_3\sim C_6$ alkynylalkyl group or a $C_3\sim C_6$ haloalkenylalkyl group,

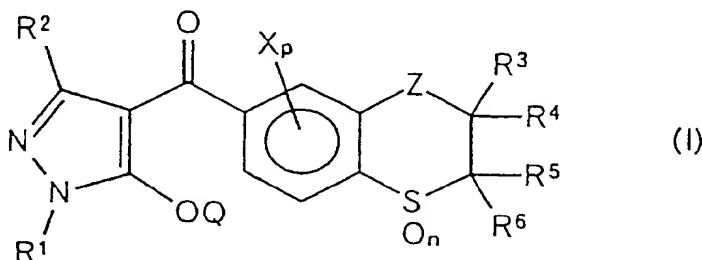
R^{12} is a $C_1\sim C_4$ alkyl group, a $C_1\sim C_4$ haloalkyl group, a $C_3\sim C_6$ cycloalkyl group, a $C_3\sim C_6$ alkenylalkyl group, a $C_3\sim C_6$ alkynylalkyl group or a $C_3\sim C_6$ haloalkenylalkyl group]],

or a salt thereof; and

at least one herbicide compound selected from the group consisting of chloroacetamide-containing herbicides such as alachlor, metolachlor, acetochlor and dimethenamide, etc., imidazoline-containing herbicides such as imazetapyr, etc., atrazine, cyanazine, metribuzin, linuron, metbenzuron, bentazone, dicamba, chlopyralid, 2,4-D, bromoxynil, pendimethalin, nicosulfuron, rimsulfuron, primisulfuron and pyridate.

PREFERRED EMBODIMENTS OF THE INVENTION

The pyrazole derivative as a first active ingredient of the herbicide composition of the present invention has the following general formula (I).



The herbicide composition of the present invention contains, as an active ingredient, at least one of pyrazole derivatives of the above general formula (I).

In the general formula (I), R^1 is a $C_1\sim C_4$ alkyl group, a $C_2\sim C_4$ alkenyl group or a $C_2\sim C_4$ haloalkenyl group, and preferred is a $C_1\sim C_4$ alkyl group. Specific examples of the $C_1\sim C_4$ alkyl group include methyl, ethyl, propyl and butyl, and the propyl and the butyl may be any one of linear, cyclic and branched ones. The $C_1\sim C_4$ alkyl group is preferably methyl or ethyl. Specific examples of the $C_2\sim C_4$ alkenyl group include $-\text{CH}=\text{CH}_2$, $-\text{CH}_2\text{CH}=\text{CH}_2$ and $-\text{CH}=\text{CH}-\text{CH}=\text{CH}_2$. The $C_2\sim C_4$ haloalkenyl group includes those in which at least one hydrogen atom of the above $C_2\sim C_4$ alkenyl group is replaced with a halogen atom (e.g., a chlorine atom, a fluorine atom, a bromine atom or an iodine atom).

In the general formula (I), R^2 is a hydrogen atom, a $C_1\sim C_4$ alkyl group, a $C_1\sim C_4$ haloalkyl group or a $C_2\sim C_4$ alkoxyalkyl group, and preferred is a hydrogen atom or a $C_1\sim C_4$ alkyl group. Specific examples of the $C_1\sim C_4$ alkyl group are as described concerning R^1 , and methyl is preferred. The $C_1\sim C_4$ haloalkyl group includes those in which at least one hydrogen atom of the $C_1\sim C_4$ alkyl group is replaced with a halogen atom (e.g., chlorine atom, a fluorine atom, a bromine atom or an iodine atom), and examples thereof include $-\text{CF}_3$, $-\text{C}_2\text{F}_5$, $-\text{C}_2\text{H}_4\text{F}$, $-\text{CH}_2\text{Cl}$, $-\text{CHF}_2$, $-\text{CCl}_3$, $-\text{C}_2\text{H}_3\text{Cl}_2$ and $-\text{C}_2\text{H}_3\text{F}_2$. Specific examples of the $C_2\sim C_4$ alkoxyalkyl group include $-\text{CH}_2\text{OCH}_3$, $-\text{CH}_2\text{OC}_2\text{H}_5$, $-\text{CH}_2\text{OC}_3\text{H}_7$, $-\text{CH}(\text{CH}_3)\text{OCH}_3$, $-\text{CH}(\text{CH}_3)\text{OC}_2\text{H}_5$, $-\text{CH}_2\text{CH}_3\text{OCH}_3$ and $-\text{CH}_2\text{CH}_2\text{OC}_2\text{H}_5$.

In the general formula (I), X is a $C_1\sim C_4$ alkyl group, a $C_1\sim C_4$ haloalkyl group, a $C_2\sim C_4$ alkoxyalkyl group, a halogen atom, a $C_1\sim C_4$ alkoxy group or a $C_1\sim C_4$ haloalkoxy group, and preferred is a $C_1\sim C_4$ alkyl group or a halogen atom. Specific examples of the $C_1\sim C_4$ alkyl group, the $C_1\sim C_4$ haloalkyl group and the $C_2\sim C_4$ alkoxyalkyl group are as described concerning R^1 or R^2 . Specific examples of the $C_1\sim C_4$ alkyl group preferably include methyl or ethyl. Specific examples of the halogen atom include a chlorine atom, a fluorine atom, a bromine atom and an iodine atom, and a chlorine atom is preferred. Specific examples of the $C_1\sim C_4$ alkoxy group include methoxy, ethoxy, propoxy and butoxy, and the propoxy and the butoxy may be linear, cyclic or branched ones. The $C_1\sim C_4$ haloalkoxy group includes those in which at least one hydrogen atom of the $C_1\sim C_4$ alkoxy group is replaced with a halogen atom (e.g., a chlorine atom, a fluorine atom, a bromine atom or an iodine atom), and examples thereof include $-\text{OCF}_3$, $-\text{OC}_2\text{F}_5$, $-\text{OC}_2\text{H}_4\text{F}$, $-\text{OC}_2\text{H}_4\text{Cl}$, $-\text{OCHF}_2$, $-\text{OCH}_2\text{F}$, $-\text{OCCl}_3$, $-\text{OC}_2\text{H}_3\text{Cl}_2$ and $-\text{OC}_2\text{H}_3\text{F}_2$.

In the general formula (I), p is the number of substituent(s) X, and it is an integer of 0, 1 or 2. When p is 1 or 2, the position(s) of the substituent(s) X is/are the 5-position and/or 8-position on the thiochroman ring.

In the general formula (I), each of R^3 , R^4 , R^5 and R^6 is independently a hydrogen atom, a $C_1\sim C_4$ alkyl group, a $C_1\sim C_4$ haloalkyl group or a $C_2\sim C_4$ alkoxyalkyl group. Each of these is preferably independently a hydrogen atom or a

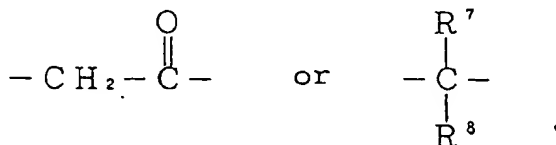
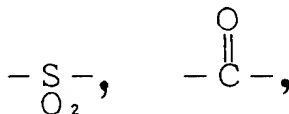
C₁~C₄ alkyl group. Specific examples of the C₁~C₄ alkyl group, the C₁~C₄ haloalkyl group and the C₂~C₄ alkoxyalkyl group are as described concerning R¹ or R².

In the general formula (I), n is the number of oxygen atom(s) bonding to the sulfur atom of the thiochroman ring, and it is an integer of 0 (sulfide), 1 (sulfoxide) or 2 (sulfone) and is preferably 2 (sulfone).

In the general formula (I), Q is a hydrogen atom or a group of -A-B.

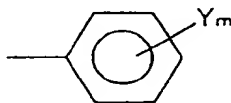
In the definition of Q,

A is a group of



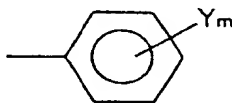
In the definition of A, each of R⁷ and R⁸ is independently a hydrogen atom or a C₁~C₄ alkyl group, and each is preferably a hydrogen atom. Specific examples of the C₁~C₄ alkyl group are as described concerning R¹.

In the definition of Q, B is a C₁~C₁₂ alkyl group, a C₃~₁₀ cycloalkyl group or a group of



Specific examples of the C₁~C₁₂ alkyl group includes those described as specific examples of the C₁~C₄ alkyl group for R¹ and others such as pentyl, hexyl, heptyl, octyl, nonyl, decanyl, undecanyl and dodecanyl. Those which have at least 3 carbon atoms may be linear or branched. Preferred is a C₁~C₈ alkyl group. Specific examples of the C₃~C₁₀ alkyl group include cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl and cycloheptyl, and cyclohexyl is preferred.

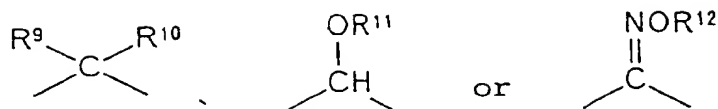
In the group of



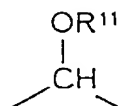
which is one embodiment of B, Y is a C₁~C₄ alkyl group, a C₁~C₄ alkoxy group, a C₁~C₄ haloalkyl group, a nitro group or a halogen atom, and preferred is a C₁~C₄ alkyl group, a C₁~C₄ alkoxy group, a nitro group or a halogen atom. Specific examples of the C₁~C₄ alkyl group, the C₁~C₄ alkoxy group, the C₁~C₄ haloalkyl group and the halogen atom are as described concerning R¹, R² or X. The C₁~C₄ alkyl group is specifically preferably methyl. The C₁~C₄ alkoxy group is specifically preferably methoxy. The halogen atom is specifically preferably a chlorine or fluorine atom.

m is the number of substituent(s) Y, and it is an integer of 0 or 1 to 3, preferably 0, 1 or 2.

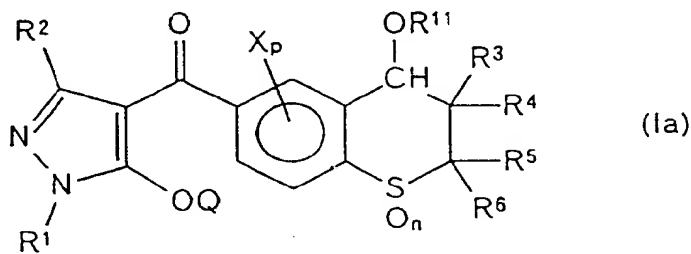
In the general formula (I), Z is



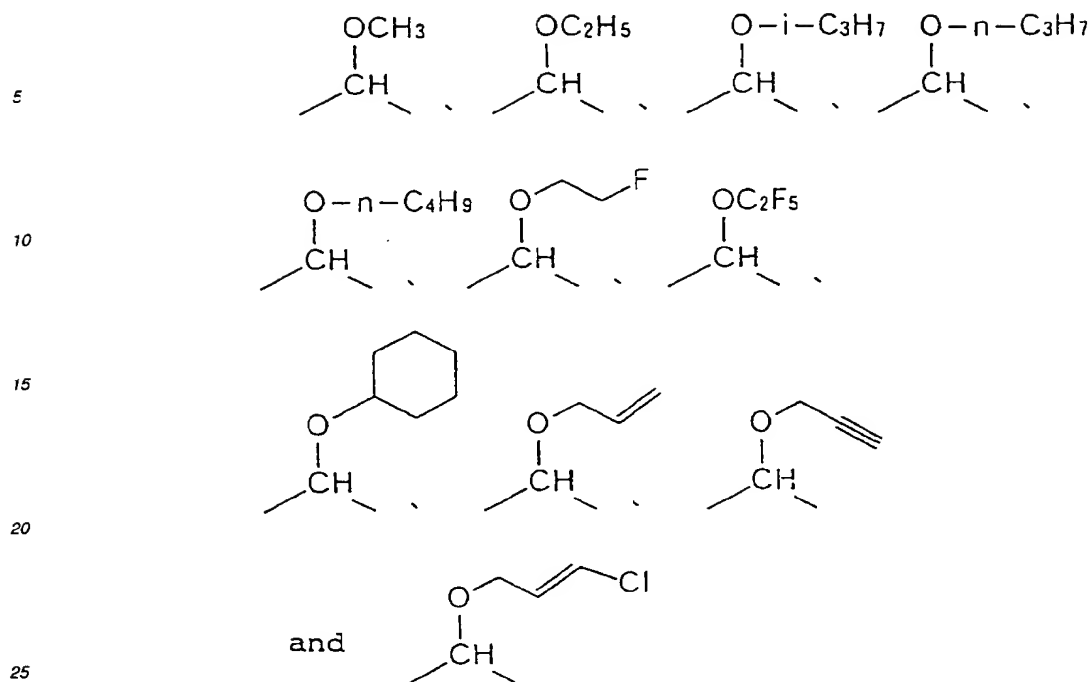
The pyrazole derivative (I) of the present invention can be classified into three kinds depending upon embodiments of Z. That is, the pyrazole derivative in which Z is



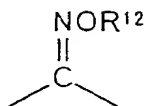
has the following general formula (Ia).



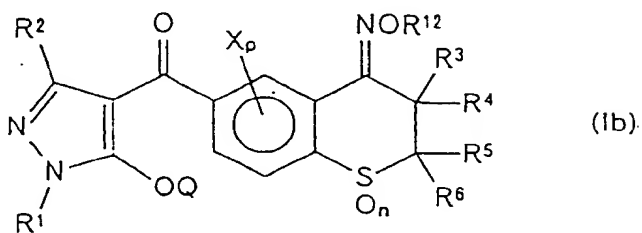
In the general formula (Ia), R^{11} is a C_1 ~ C_4 alkyl group, a C_1 ~ C_4 haloalkyl group, a C_3 ~ C_6 cycloalkyl group, a C_3 ~ C_6 alkenylalkyl group, a C_3 ~ C_6 alkynylalkyl or a C_3 ~ C_6 haloalkenylalkyl group, and preferred is a C_1 ~ C_4 alkyl group. Specific examples of the C_1 ~ C_4 alkyl group, the C_1 ~ C_4 haloalkyl group and the C_3 ~ C_6 cycloalkyl group are as described concerning R^1 , R^2 or Y. The C_1 ~ C_4 alkyl group is specifically preferably methyl or ethyl. Specific examples of the C_3 ~ C_6 alkenylalkyl group include $-CH_2-CH=CH_2$, $-CH_2CH=C(CH_3)_2$, $-CH_2C(CH_3)=CH_2$ and $-CH_2-CH=CH-C_2H_5$. Specific examples of the C_3 ~ C_6 alkynylalkyl group include $CH_2-C\equiv CH$, $-CH_2-C\equiv C-C_2H_5$ and $-CH(CH_3)-C\equiv CH$. The C_3 ~ C_6 haloalkenylalkyl group is a group in which at least one hydrogen atom of the C_3 ~ C_6 alkenylalkyl group is replaced with a halogen atom (e.g., a chlorine atom, a fluorine atom, a bromine atom or an iodine atom), and examples thereof include $-CH_2-CH=CHCl$, $-CH_2-CCl=CH_2$, $-CH_2-CF=CH_2$, $-CH_2CCl=CHCl$, $-CH_2CF=CHF$, and $-CH_2CCl=CH-CH_3$. Preferred embodiments of Z including the above R^{11} are as follows.



The pyrazole derivative in which z is

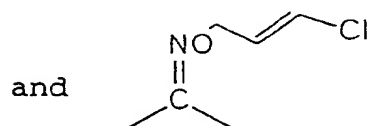
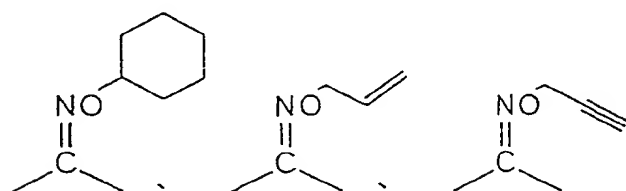
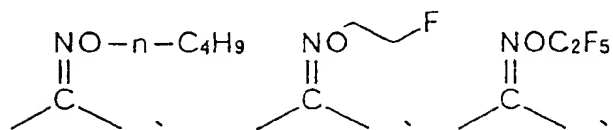
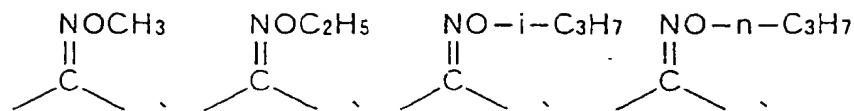


has the following general formula (Ib).

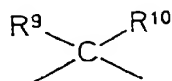


50 In the general formula (Ib), R¹² is a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group, a C₃~C₆ cycloalkyl group, a C₃~C₆ alkenylalkyl group, a C₃~C₆ alkynylalkyl group or a C₃~C₆ haloalkenylalkyl group, and a C₁~C₄ alkyl group is preferred. Specific examples of the C₁~C₄ alkyl group, the C₁~C₄ haloalkyl group, the C₃~C₆ cycloalkyl group, the C₃~C₆ alkenylalkyl group, the C₃~C₆ alkynylalkyl group and the C₃~C₆ haloalkenylalkyl group are as described concerning R¹, R², Y or R¹¹. The C₁~C₄ alkyl group is specifically preferably methyl. Preferred embodiments of Z including R¹² are as follows.

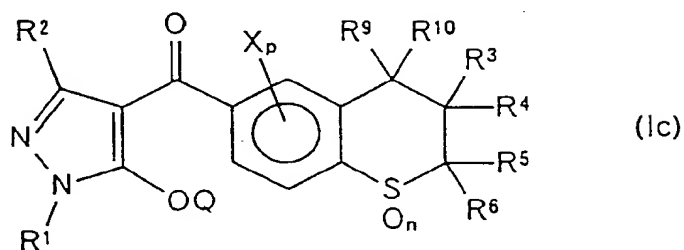
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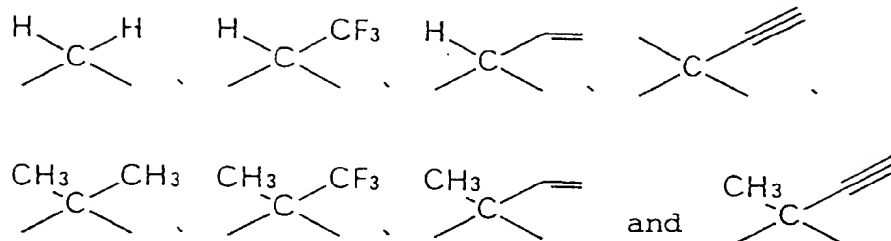
The pyrazole derivative in which Z is



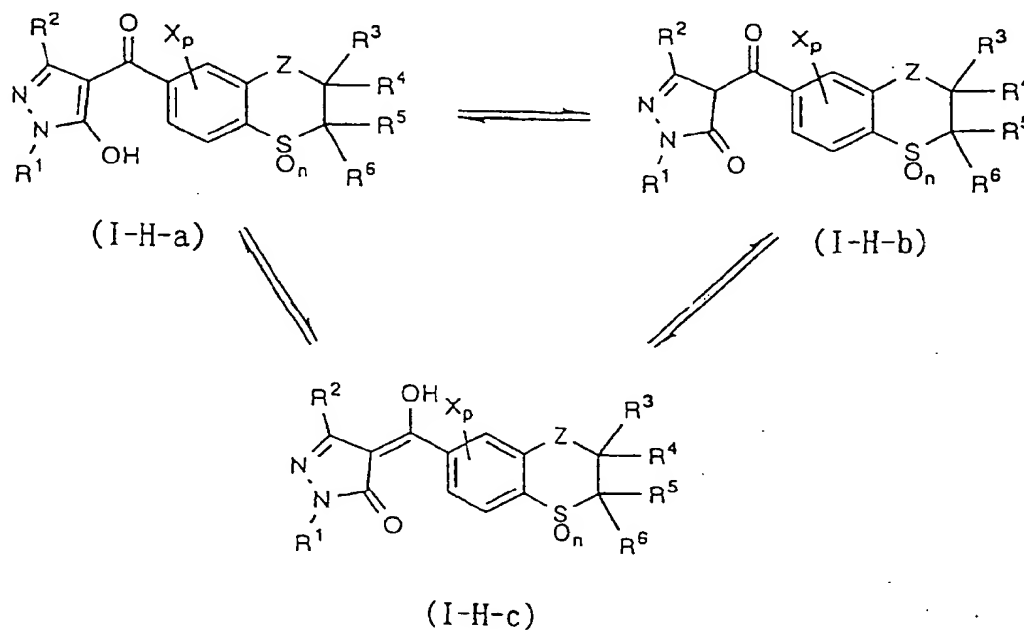
has the following general formula (Ic).



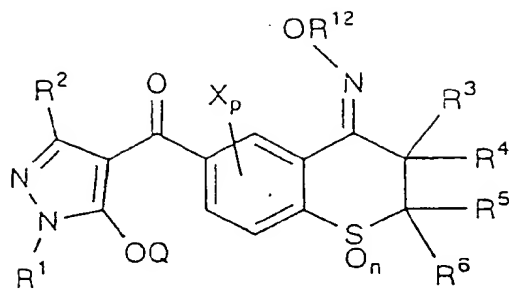
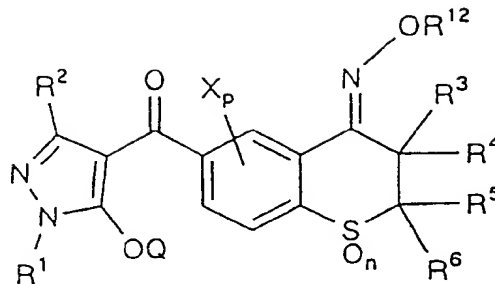
In the general formula (Ic), R^9 is a hydrogen atom, a $\text{C}_1\sim\text{C}_4$ alkyl group or a $\text{C}_1\sim\text{C}_4$ haloalkyl group, and R^{10} is a hydrogen atom, a $\text{C}_1\sim\text{C}_4$ alkyl group, a $\text{C}_2\sim\text{C}_4$ alkenyl group or a $\text{C}_2\sim\text{C}_4$ alkynyl group. Each of these is independently preferably a hydrogen atom or a $\text{C}_1\sim\text{C}_4$ alkyl group. Preferred embodiments of Z including the above R^9 and R^{10} are as follows.



15 The pyrazole derivative of the general formula (I) in which Q is a hydrogen atom includes the following three tautomers, and all of these are included in the pyrazole derivative of the present invention.



45 Further, the above pyrazole derivative (Ic) includes the following two geometrical isomers, and both of these are included in the pyrazole derivative of the present invention.

(Ic- α)(Ic- β)

Some of pyrazole derivatives of the general formula (I) have asymmetric carbon, and include a variety of isomers, and the pyrazole derivative of the present invention includes all of such isomers or mixtures of isomers.

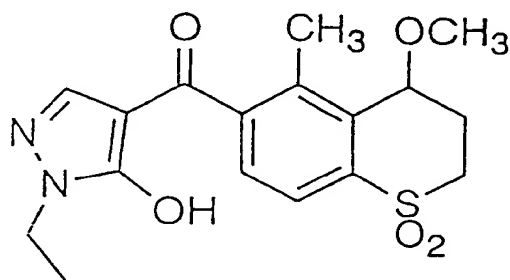
Further, the pyrazole derivative of the general formula (I) in which Q is a hydrogen atom is acidic, and can be easily converted to a salt by treating it with a base. This salt is also included in the pyrazole derivative of the present invention.

As the above base, any known base may be used without any special limitation. Examples thereof include organic bases such as amines and anilines and inorganic bases such as a sodium compound and a potassium compound. The amines include monoalkylamine, dialkylamine and trialkylamine. The alkyl group of each of the alkylamines is generally a C₁~C₄ alkyl group. The anilines include aniline, monoalkylaniline and dialkylaniline. The alkyl group of each of the alkylanilines is generally a C₁~C₄ alkyl group. The sodium compound includes sodium hydroxide and sodium carbonate. The potassium compound includes sodium hydroxide and sodium carbonate.

Preferably, specific examples, chemical names and structural formulae of pyrazole derivatives of the general formula (Ia) are as follows.

Compound(Ia-1)

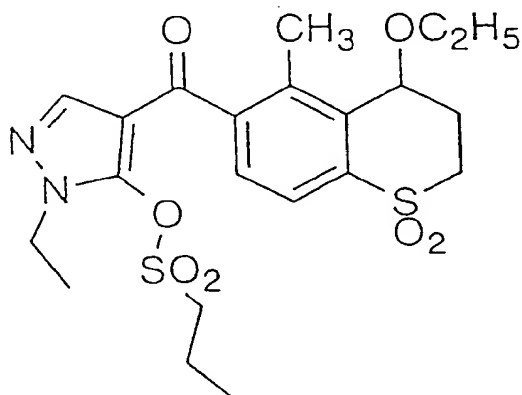
4-methoxy-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia), R¹=C₂H₅, R²=H, R³=R⁴=R⁵=R⁶=H, X=5-CH₃, R¹¹=CH₃, Q=H, n=2, p=1)

Compound (Ia-2)

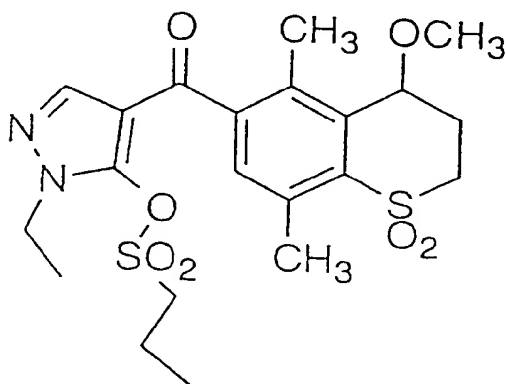
4-ethoxy-5-methyl-6-(1-ethyl-5-n-propanesulfonyloxyprazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5-CH_3$, $R^{11} = C_2H_5$, $Q = -SO_2-n-C_3H_7$, $n=2$, $p=1$)

Compound (Ia-3)

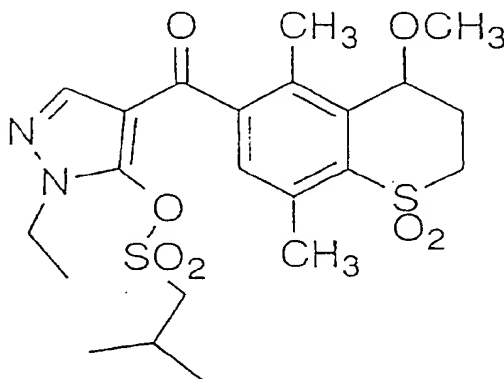
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-n-propanesulfonyloxyprazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5, 8-CH_3$, $R^{11} = CH_3$, $Q = -SO_2-n-C_3H_7$, $n=2$, $p=2$)

Compound (Ia-4)

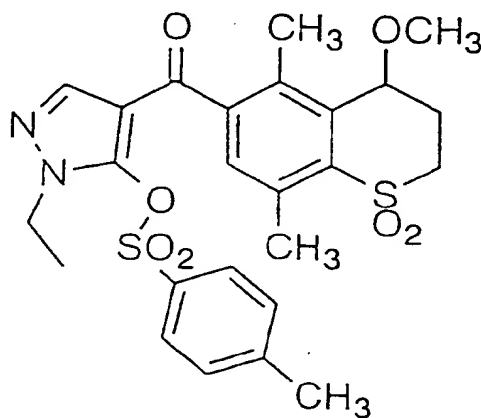
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-i-butanefonyloxy-pyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{11} = CH_3$, $Q = -SO_2-i-C_4H_9$, $n=2$, $p=2$)

Compound (Ia-5)

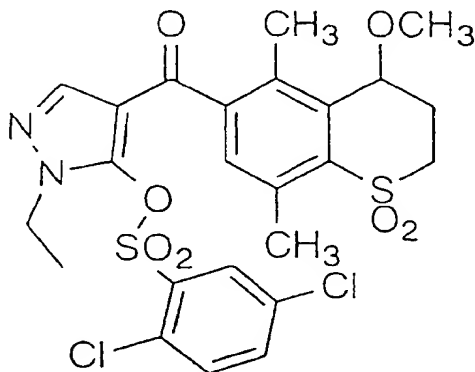
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-p-toluenesulfonyloxy-pyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{11} = CH_3$, $Q = -SO_2-C_6H_4-CH_3$, $n=2$, $p=2$)

Compound(Ia-6)

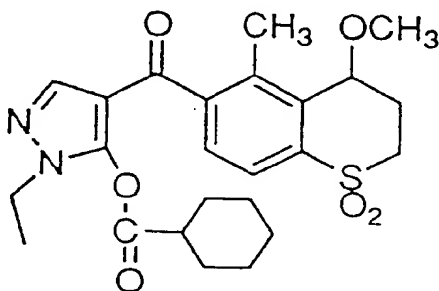
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-(2,5-dichlorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^{11}=CH_3$, $Q=-SO_2-C_6H_3-Cl_2$, $n=2$, $p=2$)

Compound(Ia-7)

4-methoxy-5-methyl-6-(1-ethyl-5-cyclohexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide

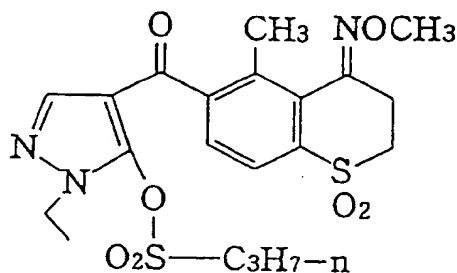


(In the general formula (Ia), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{11}=CH_3$, $Q=-CO-cyclo-C_6H_{11}$, $n=2$, $p=1$)

Preferably, specific examples, chemical names and structural formulae of pyrazole derivative of the general formula (Ib) are as follows.

Compound(Ib-1)

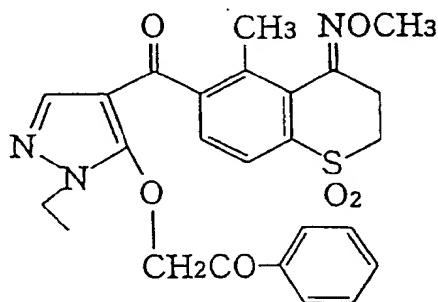
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-SO_2-n-C_3H_7$, $n=2$, $p=1$)

Compound(Ib-2)

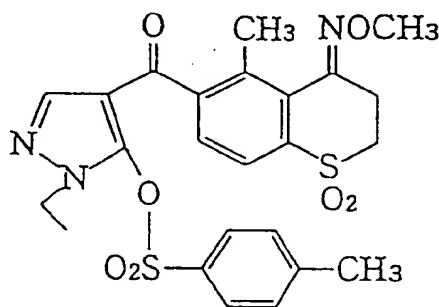
4-methoxyimino-5-methyl-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-CH_2CO-C_6H_5$, $n=2$, $p=1$)

Compound(Ib-3)

4-methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide

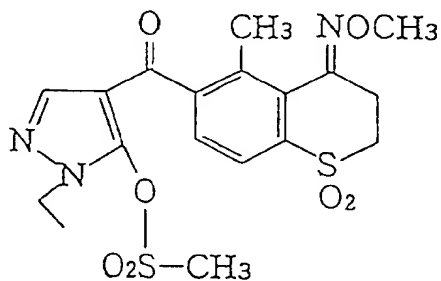


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(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-SO_2-C_6H_4-CH_3$, $n=2$, $p=1$)

Compound(Ib-4)

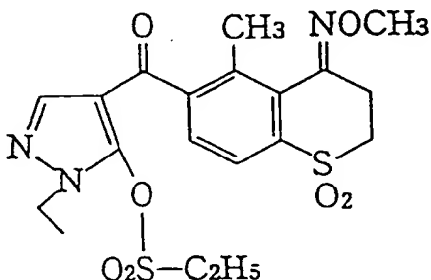
4-methoxyimino-5-methyl-6-(1-ethyl-5-methanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=SO_2-CH_3$, $n=2$, $p=1$)

Compound(Ib-5)

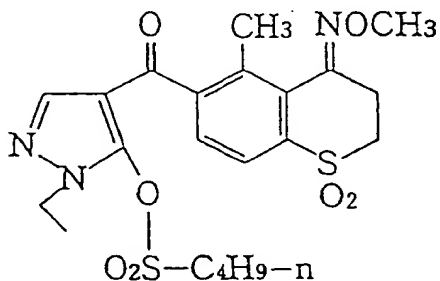
4-methoxyimino-5-methyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-SO_2-C_2H_5$, $n=2$, $p=1$)

Compound(Ib-6)

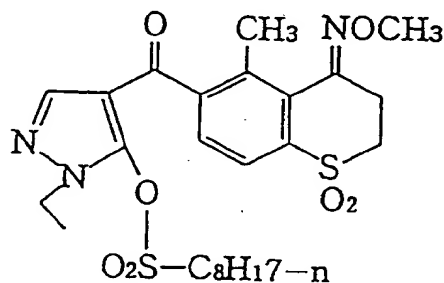
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-butanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-SO_2-n-C_4H_9$, $n=2$, $p=1$)

Compound(Ib-7)

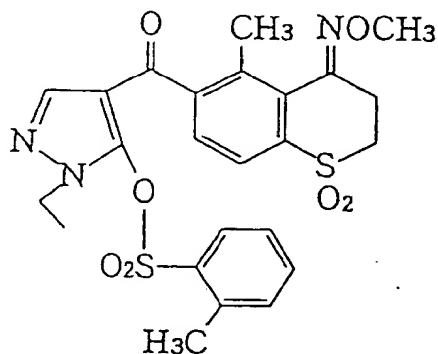
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-octanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-SO_2-n-C_8H_{17}$, $n=2$, $p=1$)

Compound (Ib-8)

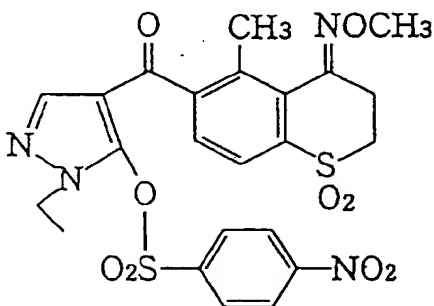
4-methoxyimino-5-methyl-6-(1-ethyl-5-(2-methylphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_4-CH_3$, $n=2$, $p=1$)

Compound (Ib-9)

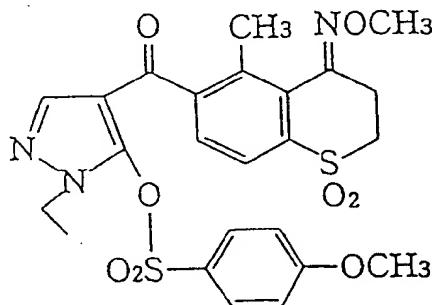
4-methoxyimino-5-methyl-6-(1-ethyl-5-(2-nitrophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_4-NO_2$, $n=2$, $p=1$)

Compound (Ib-10)

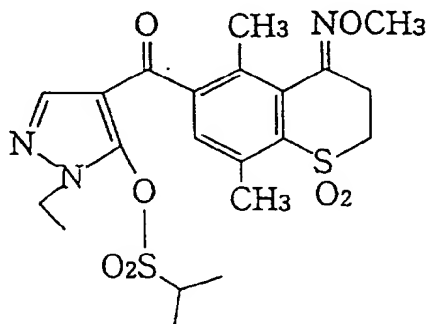
4-methoxyimino-5-methyl-6-(1-ethyl-5-(4-methoxyphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_4-OCH_3$, $n=2$, $p=1$)

Compound (Ib-11)

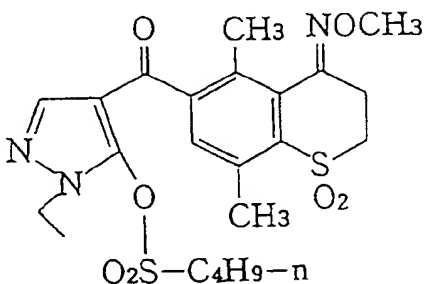
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-i-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5, 8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-i-C_3H_7$, $n=2$, $p=2$)

Compound (Ib-12)

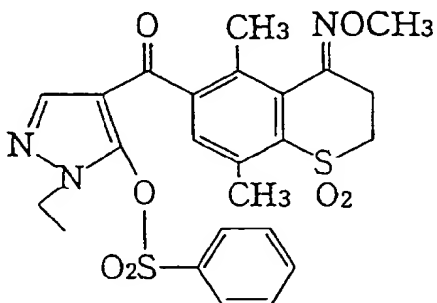
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-n-butanefonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-n-C_4H_9$, $n=2$, $p=2$)

Compound (Ib-13)

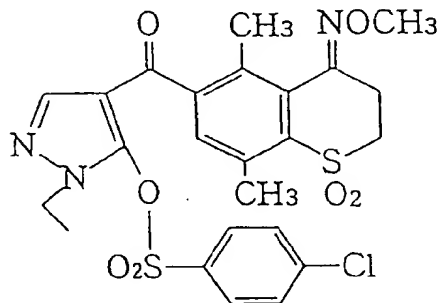
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-phenylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_5$, $n=2$, $p=2$)

Compound(Ib-14)

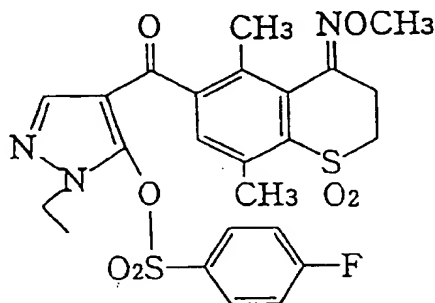
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-(4-chlorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_4-Cl$, $n=2$, $p=2$)

Compound(Ib-15)

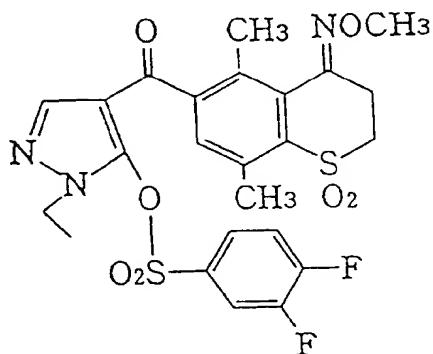
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-(4-fluorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_4-F$, $n=2$, $p=2$)

Compound (Ib-16)

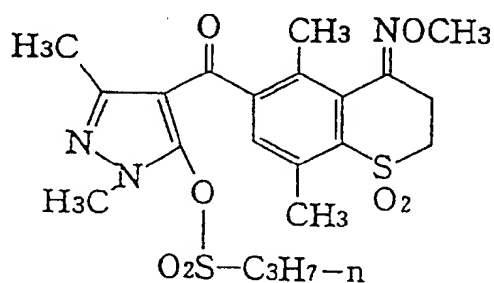
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-(3,4-difluorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-C_6H_3-F_2$, $n=2$, $p=2$)

Compound (Ib-17)

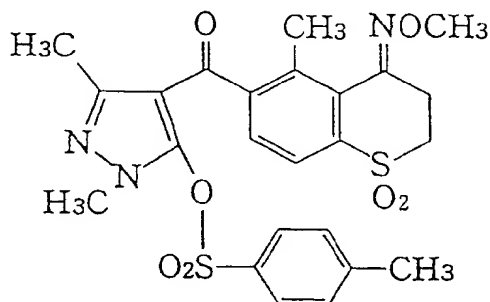
5,8-dimethyl-4-methoxyimino-(1,3-dimethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^{12} = CH_3$, $Q = -SO_2-n-C_3H_7$, $n=2$, $p=2$)

Compound(Ib-18)

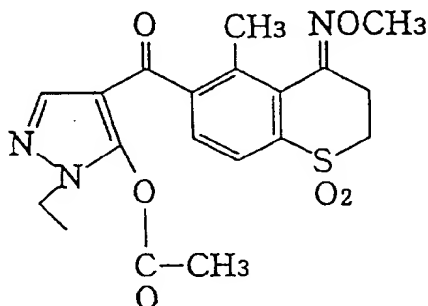
4-methoxyimino-5-methyl-(1,3-dimethyl-5-p-toluensulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=CH_3$, $R^2=CH_3$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=SO_2-C_6H_4-CH_3$, $n=2$, $p=1$)

Compound(Ib-19)

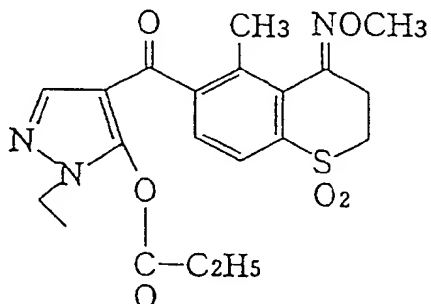
4-methoxyimino-5-methyl-6-(1-ethyl-5-acetyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=CO-CH_3$, $n=2$, $p=1$)

Compound (Ib-20)

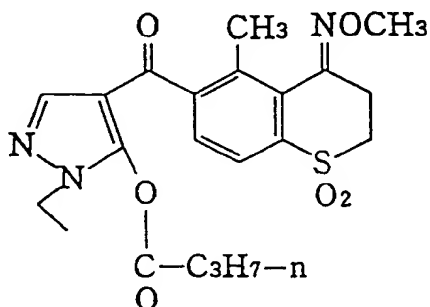
4-methoxyimino-5-methyl-6-(1-ethyl-5-ethanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5-CH_3$, $R^{12} = CH_3$, $Q = -CO-C_2H_5$, $n=2$, $p=1$)

Compound (Ib-21)

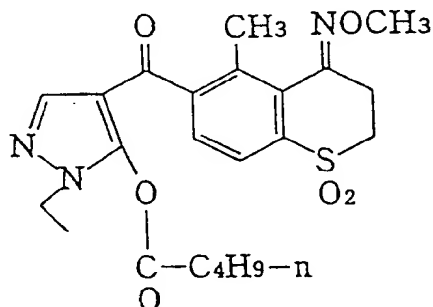
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5-CH_3$, $R^{12} = CH_3$, $Q = -CO-n-C_3H_7$, $n=2$, $p=1$)

Compound(Ib-22)

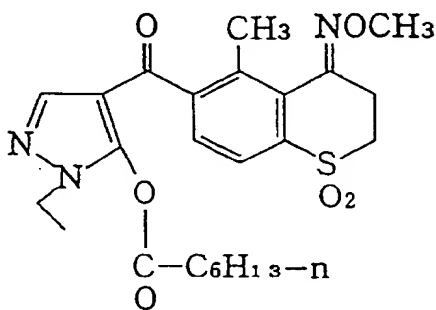
4-methoxyimino-5-metyl-6-(1-ethyl-5-n-butanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-CO-n-C_4H_9$, $n=2$, $p=1$)

Compound(Ib-23)

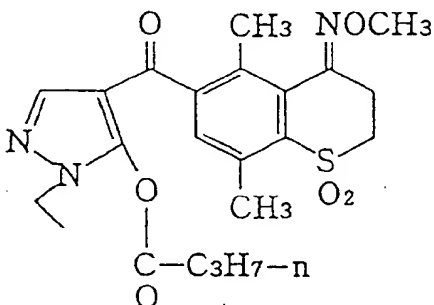
4-methoxyimino-5-metyl-6-(1-ethyl-5-n-hexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-CO-n-C_6H_{13}$, $n=2$, $p=1$)

Compound(Ib-24)

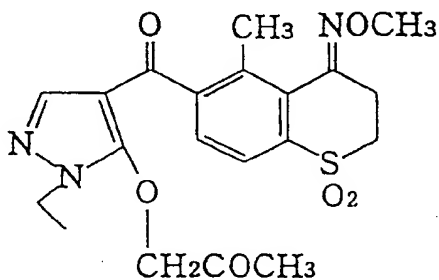
5,8-dimethyl-4-methoxyimino-6-(1-ethyl-5-n-propanecarbonyloxy-pyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^{12}=CH_3$, $Q=-CO-n-C_3H_7$, $n=2$, $p=2$)

Compound(Ib-25)

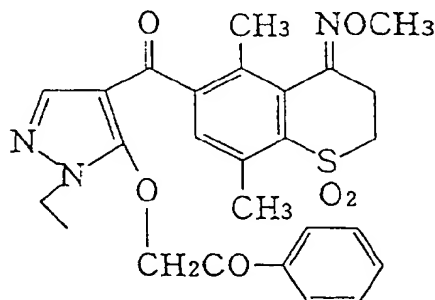
4-methoxyimino-5-methyl-6-(1-ethyl-5-acetylmethyleneoxy-pyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-CH_3$, $R^{12}=CH_3$, $Q=-CH_2COCH_3$, $n=2$, $p=1$)

Compound(Ib-26)

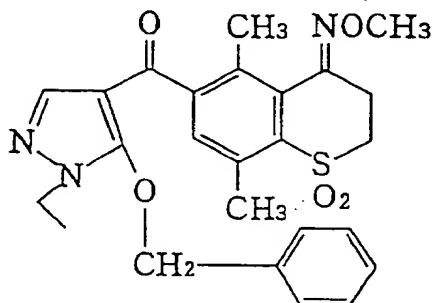
5,8-dimethyl-4-methoxyimino-6-(1-ethyl-5-phenacyloxy-pyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^{12}=CH_3$, $Q=CH_2CO-C_6H_5$, $n=2$, $p=2$)

Compound(Ib-27)

5,8-dimethyl-4-methoxyimino-6-(1-ethyl-5-benzyloxy-pyrazol-4-yl)carbonylthiochroman-1,1-dioxide

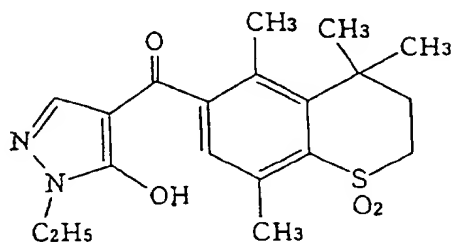


(In the general formula (Ib), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^{12}=CH_3$, $Q=CH_2-C_6H_5$, $n=2$, $p=2$)

Preferably, specific examples, chemical names and structural formulae of pyrazole derivative of the general formula (Ic) are as follows.

Compound(Ic-1)

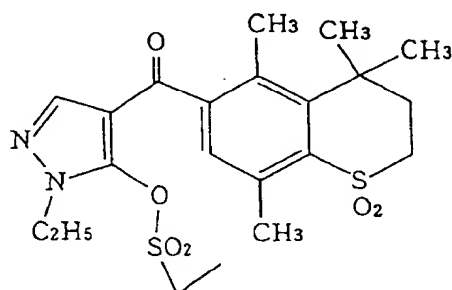
4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=2$)

Compound(Ic-2)

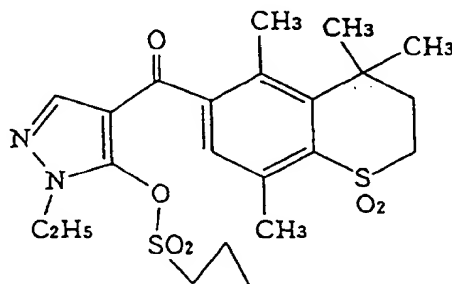
4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-C_2H_5$, $n=2$, $p=2$)

Compound(Ic-3)

4,4,5,8-tetramethyl-6-(1-ethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide

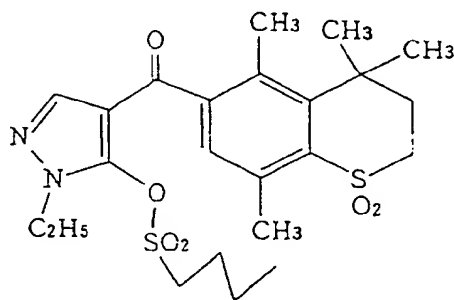


(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-n-C_3H_7$,

n=2, p=2)

Compound(Ic-4)

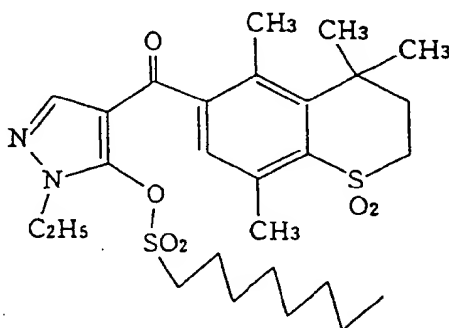
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-butanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^9 = R^{10} = CH_3$, $Q = -SO_2-n-C_4H_9$, $n=2$, $p=2$)

Compound(Ic-5)

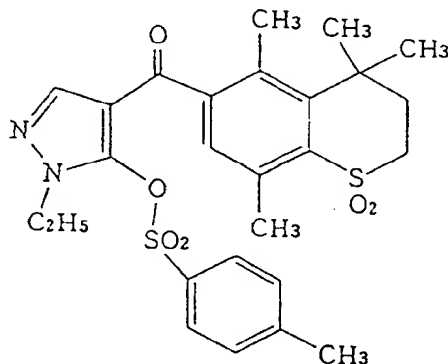
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-octanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^9 = R^{10} = CH_3$, $Q = -SO_2-C_8H_{17}$, $n=2$, $p=2$)

Compound(Ic-6)

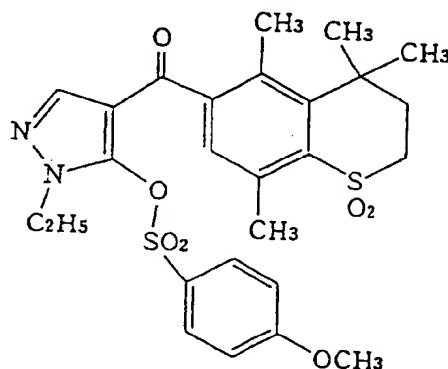
4,4,5,8-tetramethyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^9 = R^{10} = CH_3$, $Q = -SO_2-C_6H_4-CH_3$, $n=2$, $p=2$)

Compound(Ic-7)

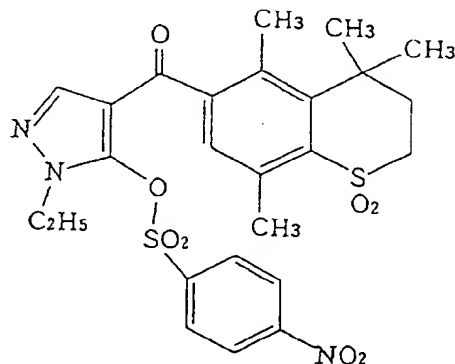
4,4,5,8-tetramethyl-6-(1-ethyl-5-(4-methoxyphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^9 = R^{10} = CH_3$, $Q = -SO_2-C_6H_4-OCH_3$, $n=2$, $p=2$)

Compound(Ic-8)

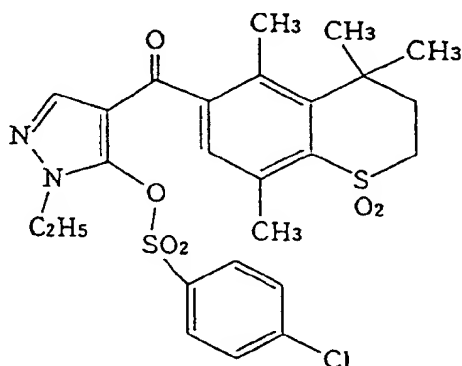
4,4,5,8-tetramethyl-6-(1-ethyl-5-(4-nitrophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-C_6H_4-NO_2$, $n=2$, $p=2$)

Compound(Ic-9)

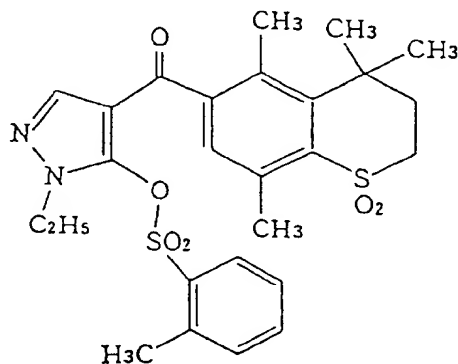
4,4,5,8-tetramethyl-6-(1-ethyl-5-p-chlorophenylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-C_6H_4-Cl$, $n=2$, $p=2$)

Compound(Ic-10)

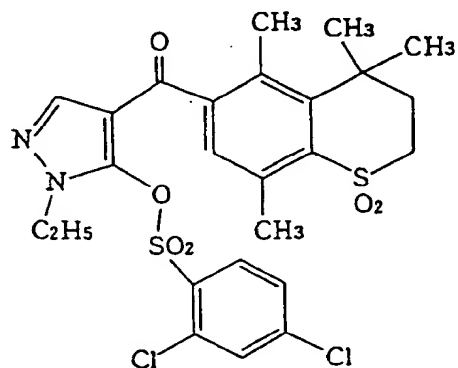
4,4,5,8-tetramethyl-6-(1-ethyl-5-(2-methylphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-C_6H_4-CH_3$, $n=2$, $p=2$)

Compound(Ic-11)

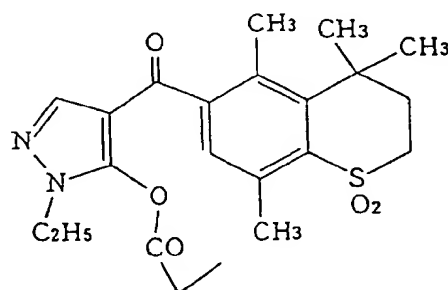
4,4,5,8-tetramethyl-6-(1-ethyl-5-(2,4-dichlorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-C_6H_3-Cl$, $n=2$, $p=2$)

Compound(Ic-12)

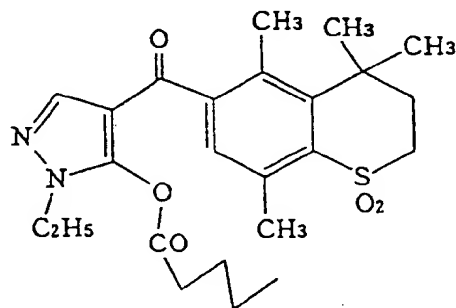
4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-CO-C_2H_5$, $n=2$, $p=2$)

Compound(Ic-13)

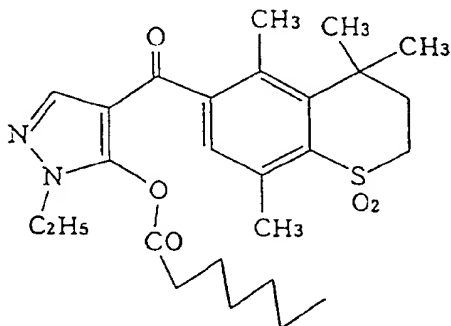
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-butanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-CO-n-C_4H_9$, $n=2$, $p=2$)

Compound(Ic-14)

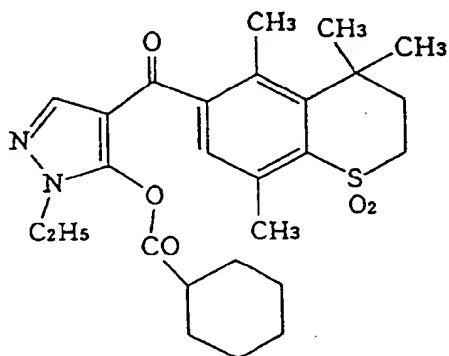
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-hexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, 8- CH_3 , $R^9 = R^{10} = CH_3$, $Q = -CO-n-C_6H_{13}$, $n=2$, $p=2$)

Compound(Ic-15)

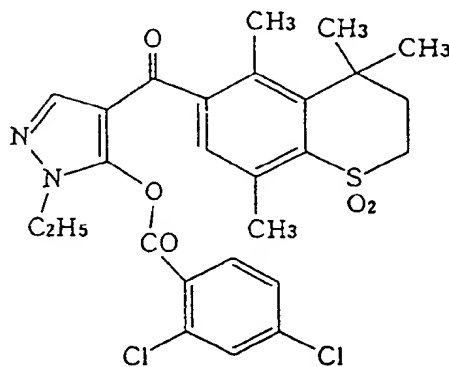
4,4,5,8-tetramethyl-6-(1-ethyl-5-cyclohexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, 8- CH_3 , $R^9 = R^{10} = CH_3$, $Q = -CO-cyclo-C_6H_{11}$, $n=2$, $p=2$)

Compound(Ic-16)

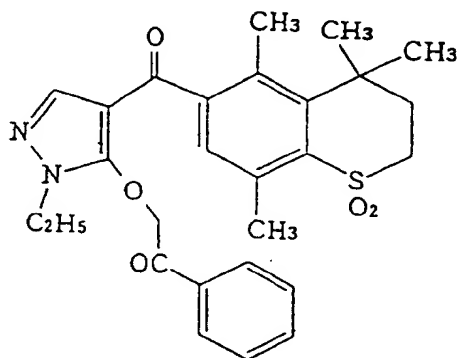
4,4,5,8-tetramethyl-6-(1-ethyl-5-(2,4-dichlorophenyl)carbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^9 = R^{10} = CH_3$, $Q = -CO-C_6H_4-Cl_2$, $n=2$, $p=2$)

Compound(Ic-17)

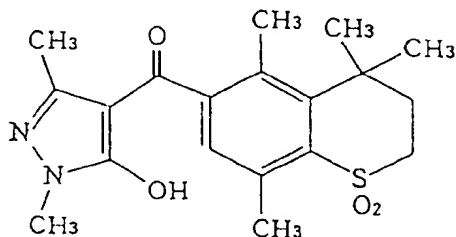
4,4,5,8-tetramethyl-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1 = C_2H_5$, $R^2 = H$, $R^3 = R^4 = R^5 = R^6 = H$, $X = 5$, $8-CH_3$, $R^9 = R^{10} = CH_3$, $Q = -CO-C_6H_5$, $n=2$, $p=2$)

Compound(Ic-18)

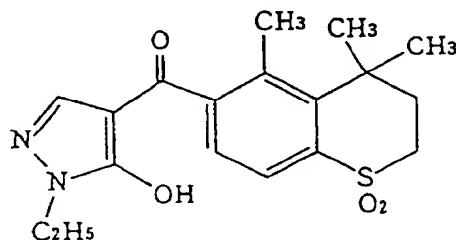
4,4,5,8-tetramethyl-(1,3-dimethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=CH_3$, $R^2=CH_3$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=2$)

Compound(Ic-19)

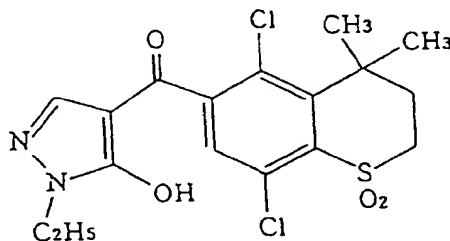
4,4,5-trimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=1$)

Compound(Ic-20)

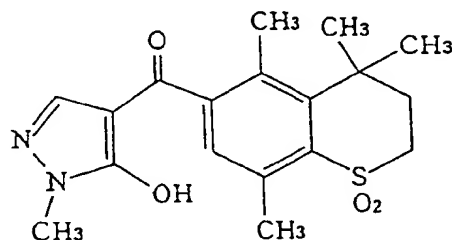
5,8-dichloro-4,4-dimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-Cl$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=2$)

Compound(Ic-21)

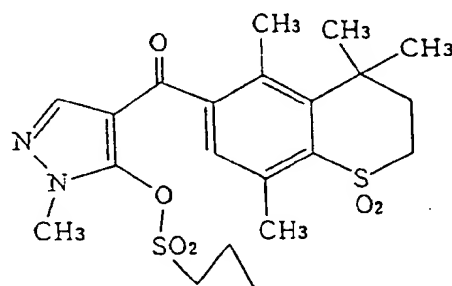
4,4,5,8-tetramethyl-6-(1-methyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=CH_3$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=2$)

Compound(Ic-22)

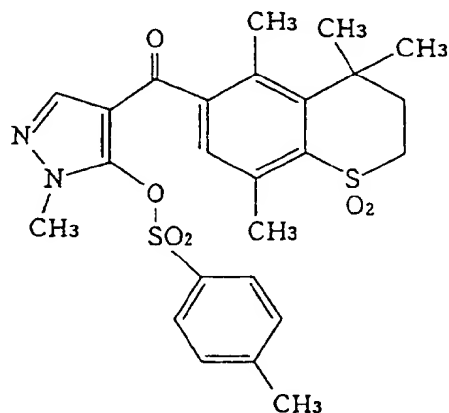
4,4,5,8-tetramethyl-6-(1-methyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=CH_3$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-n-C_3H_7$, $n=2$, $p=2$)

Compound(Ic-23)

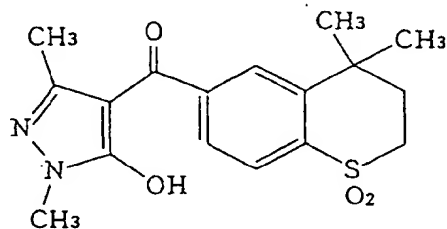
4,4,5,8-tetramethyl-6-(1-methyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(in the general formula (Ic), $R^1=CH_3$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=-SO_2-C_6H_4-CH_3$, $n=2$, $p=2$)

Compound(Ic-24)

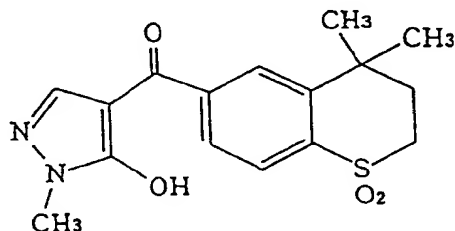
4,4-dimethyl-(1,3-dimethyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=CH_3$, $R^2=CH_3$, $R^3=R^4=R^5=R^6=H$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=0$)

Compound(Ic-25)

4,4-dimethyl-(1-methyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide

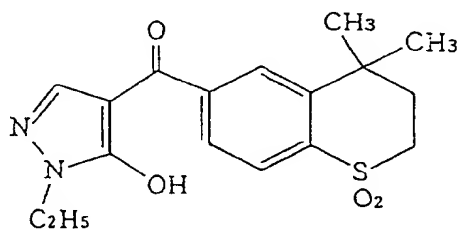


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(In the general formula (Ic), $R^1=CH_3$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=0$)

Compound(Ic-26)

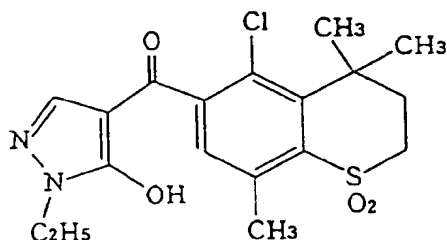
4,4-dimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=0$)

Compound(Ic-27)

5-chloro-4,4,8-trimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic), $R^1=C_2H_5$, $R^2=H$, $R^3=R^4=R^5=R^6=H$, $X=5-Cl$, $8-CH_3$, $R^9=R^{10}=CH_3$, $Q=H$, $n=2$, $p=2$)

In the herbicide composition of the present invention, the herbicide compound which is the second active ingredient can exhibit a synergistic effect when used with the above mentioned pyrazole derivative (I). Examples of this herbicide compound includes germination inhibitor such as chloroacetamide herbicide or dinitroaniline herbicide; plant hormone such as benzoic acid herbicide, pyridine carboxylic acid herbicide or phenoxy herbicide; ALS (Acetolactate Synthetase) inhibitor such as sulfonylurea herbicide or imidazolinone herbicide; photosynthesis inhibitor such as triazine herbicide or urea herbicide; and other herbicide such as diazine herbicide, benzonitrile herbicide or pyridazine herbicide.

In the herbicide composition of the present invention, among the above mentioned herbicide compound, at least one member selected from the following groups is used as an active ingredient with the pyrazole derivative (I).

Chloroacetamide herbicide;

Imidazolinone herbicide;

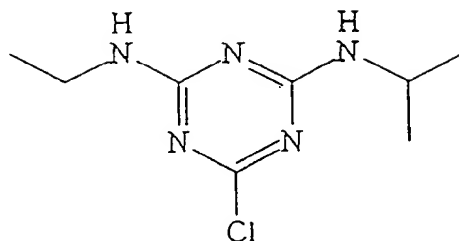
Triazine herbicide such as Compound (B-1) ~ (B-3);

Compound(B-1)

Common name:atrazine

Chemical Name:

6-chloro-N²-ethyl-N⁴-isopropil-1,3,5-triazine-2,4-diamine

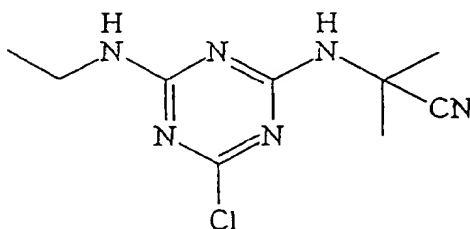


Compound(B-2)

Common name: cyanazine

Chemical Name:

2-(4-chloro-6-ethylamino-1,3,5-triazin-2-ylamino)-2-methylpropionitrile

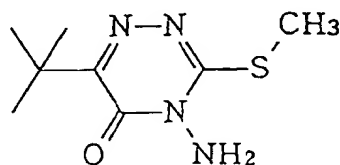


Compound(B-3)

Common name: metribuzin

Chemical Name:

4-amino-6-tert-butyl-4,5-dihydro-3-methylthio-1,2,4-triazin-5-one



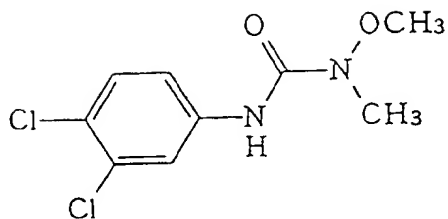
Urea herbicide such as Compounds (B-4) and (B-5) ;

Compound(B-4)

Common name: linuron

Chemical Name:

3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea

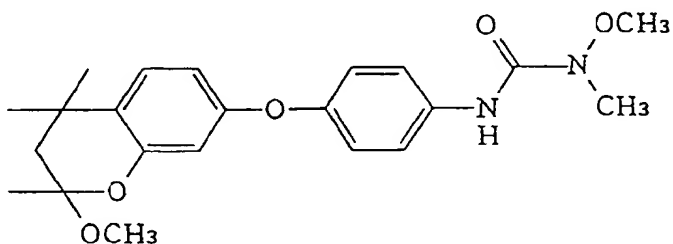


Compound(B-5)

Common name: metbenzuron

Chemical Name:

(±)-1-methoxy-3-[4-(2-methoxy-2,4,4-trimethylchroman-7-yloxy)phenyl]-1-methylurea



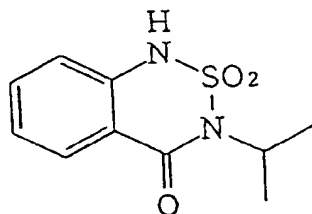
Diazine herbicide such as Compound (B-6) ;

Compound(B-6)

Common name: bentazone

Chemical Name:

3-isopropyl-1H-2,1,3-benzothiadiazin-4(3H)-one-2,2-dioxide



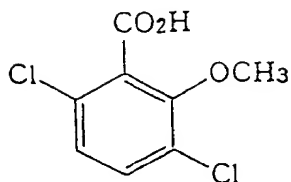
Benzoic acid herbicide such as Compound (B-7);

Compound(B-7)

Common name: dicamba

Chemical Name:

3,6-dichloro-2-methoxybenzoic acid



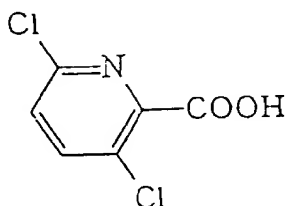
Pyridine carboxylic acid herbicide such as Compound (B-8) ;

Compound(B-8)

Common name: chlopyralid

Chemical Name:

3,6-dichloropyridine-2-carboxylic acid



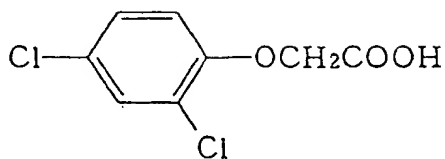
Phenoxy herbicide such as Compound (B-9) ;

Compound(B-9)

Common name: 2,4-D

Chemical Name:

2-(2,4-dichlorophenoxy)acetic acid



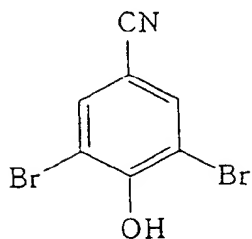
Benzonitrile herbicide such as Compound (B-10) ;

Compound(B-10)

Common name: bromoxynil

Chemical Name:

3,5-dibromo-4-hydroxybenzonitrile



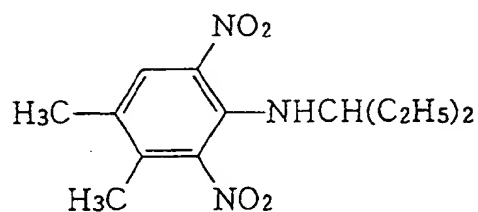
Dinitroaniline herbicide such as Compound (B-13) ;

Compound(B-13)

Common name: pendimethalin

Chemical Name:

N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine



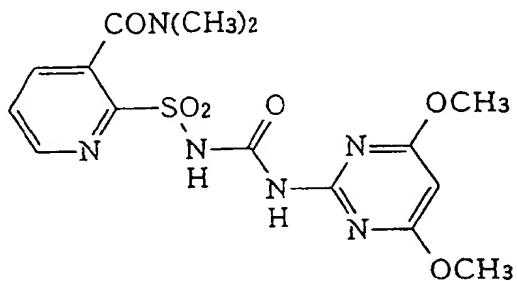
Sulfonyl urea herbicide such as Compound (B-14), (B-15) and (B-17) ;

Compound(B-14)

Common name: nicosulfuron

Chemical Name:

2-(4,6-dimethoxypyrimidin-2-yl)carbamoylsulfamoyl)-N,N-dimethylnicotineamide

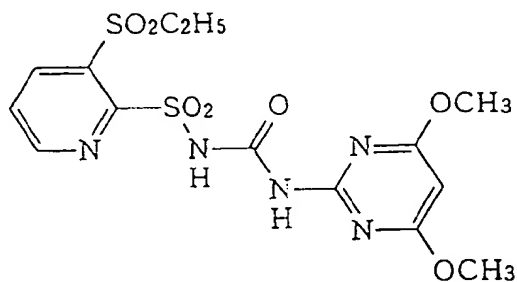


Compound(B-15)

Common name: rimsulfuron

Chemical Name:

1-(4,6-dimethoxypyrimidin-2-yl-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea

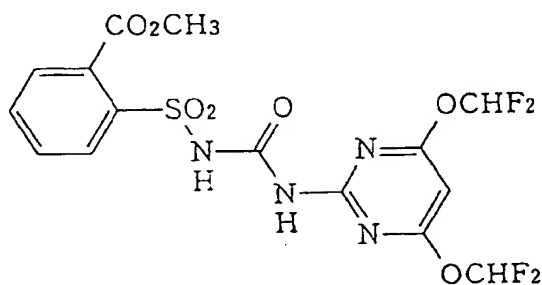


Compound(B-17)

Common name:primisulfuron

Chemical Name:

Methyl 2-[4,6-bis(difluoromethoxy)pyrimidin-2-yl-carbamoylsulfamoyl]benzoic acid



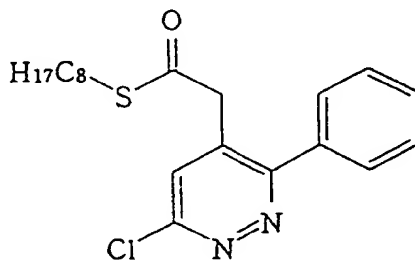
Pyridazin herbicide such as Compound (B-20) ;

Compound(B-20)

Common name:pyridate

Chemical Name:

6-chloro-3-phenylpyridazin-4-yl-S-octylthiocarbonate

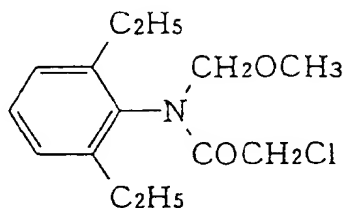


Among the herbicide compounds, the following Compounds (B-11), (B-12), (B-18) and (B-19) are exemplified as

the chloroacetamide herbicide.

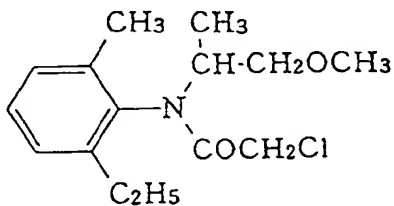
Compound(B-11)

5 Common name:alachlor
Chemical Name:
2-chloro-2',6'-diethyl-N-methoxymethylacetanilide



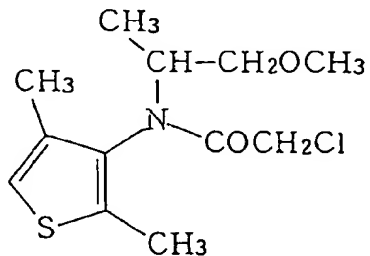
20 Compound(B-12)

Common name:metolachlor
Chemical Name:
2-chloro-N-(2-ethyl-6-methylphenyl)-N-(2-methoxy-1-methylethyl)acetamide



Compound(B-18)

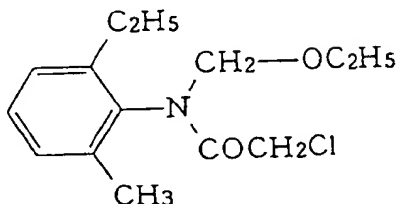
40 Common name:dimethenamid
Chemical Name:
(1RS,aRS)-2-chloro-N-(2,4-dimethyl-3-thienyl)-N-(2-methoxy-1-methylethyl)acetamide



Compound(B-19)

Common name:acetochlor
Chemical Name:

2-chloro-2'-ethyl-6'-methyl-N-ethoxymethylacetanilide



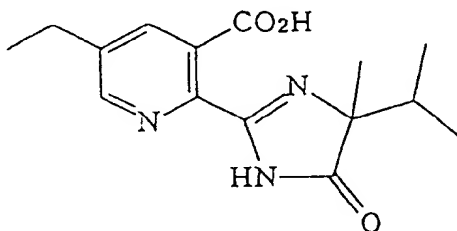
Among the herbicide compounds, the following Compounds (B-16) and (B-21) are exemplified as the imidazolinone herbicide.

Compound(B-16)

Common name: imazethapyr

Chemical Name:

5-ethyl-2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)nicotinic acid

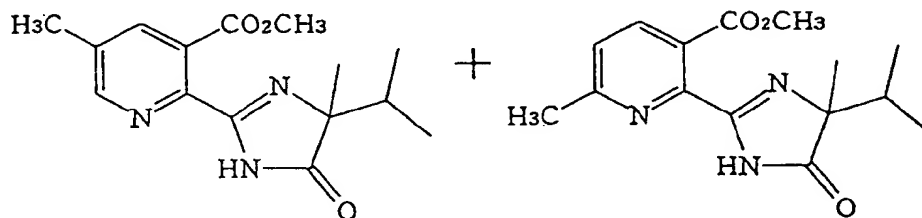


Compound(B-21)

Common name: imazamethabenz-methyl

Chemical Name:

Compound of a Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-m-toluic acid and a Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-p-toluic acid



(Imazamethabenz-methyl which is a mixture of the two isomers is commercially available under a trade name of Assert.)

The herbicide composition of the present invention contains, as active ingredients, at least one of the above pyrazole derivatives (I) and at least one selected from a group of the above herbicide compounds. The mixing ratio of these is not specially limited, and a synergistic effect can be obtained in a broad range of the mixing ratio, while the pyrazole derivative (I) and the compounds (B-1)~(B-2) are generally preferably mixed in the following mixing ratios (weight

ratios).

pyrazole derivative(I):compound(B-1:atrazine)=2:1~1:50
 pyrazole derivative(I):compound(B-2:cyanazine)=2:1~1:50
 5 pyrazole derivative(I):compound(B-3:metribuzin)=3:1~1:25
 pyrazole derivative(I):compound(B-4:linuron)=2:1~1:50
 pyrazole derivative(I):compound(B-5:metbenzuron)=1:2~ 1:100
 pyrazole derivative(I):compound(B-6:bentazone)=6:1~1:100
 pyrazole derivative(I):compound(B-7:dicamba)=1:1~1:50
 10 pyrazole derivative(I):compound(B-8:chlorypyridid)=4:3~1:12
 pyrazole derivative(I):compound(B-9:2,4-D)=2:1~1:5
 pyrazole derivative(I):compound(B-10:bromoxynil)=1:1~1:50
 pyrazole derivative(I):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(I):compound(B-12:metolachlor)=2:1~1:25
 15 pyrazole derivative(I):compound(B-13:pendimethalin)=2:1~ 1:25
 pyrazole derivative(I):compound(B-14:nicosulfuron)=1:3~ 40:1
 pyrazole derivative(I):compound(B-15:rimsulfuron)=1:3~40:1
 pyrazole derivative(I):compound(B-16:imazethapyr)=1:6~40:1
 pyrazole derivative(I):compound(B-17:primisulfuron)=1:3~ 40:1
 20 pyrazole derivative(I):compound(B-18:dimethenamid)=2:1~ 1:50
 pyrazole derivative(I):compound(B-19:acetochlor)=2:1~1:50
 pyrazole derivative(I):compound(B-20:pyridate)=3:2~1:50
 pyrazole derivative(I):compound(B-21:imazamethabenzmethyl)=1:6~40:1

25 The above pyrazole derivatives (Ia), (Ib) and (Ic) and the compounds (B-1)~(B-20) are preferably mixed in the following mixing ratios, respectively.

pyrazole derivative(Ia):compound(B-1:atrazine)=1:1~1:50
 pyrazole derivative(Ia):compound(B-2:cyanazine)=2:1~1:25
 30 pyrazole derivative(Ia):compound(B-3:metribuzin)=3:1~1:12
 pyrazole derivative(Ia):compound(B-4:linuron)=2:1~1:25
 pyrazole derivative(Ia):compound(B-6:bentazone)=1:2~1:100
 pyrazole derivative(Ia):compound(B-7:dicamba)=1:1~1:50
 pyrazole derivative(Ia):compound(B-9:2,4-D)=2:1~1:50
 35 pyrazole derivative(Ia):compound(B-10:bromoxynil)=1:1~1:50
 pyrazole derivative(Ia):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(Ia):compound(B-12:metolachlor)=2:1~ 1:25
 pyrazole derivative(Ia):compound(B-13:pendimethalin)=2:1~ 1:25
 pyrazole derivative(Ia):compound(B-14:nicosulfuron)=1:3~ 40:1
 40 pyrazole derivative(Ia):compound(B-15:rimsulfuron)=1:3~ 40:1
 pyrazole derivative(Ia):compound(B-16:imazethapyr)=1:6~ 40:1
 pyrazole derivative(Ia):compound(B-17:primisulfuron)=1:3~ 40:1
 pyrazole derivative(Ia):compound(B-18:dimethenamid)=2:1~ 1:50
 pyrazole derivative(Ia):compound(B-19:acetochlor)=2:1~1:50
 45 pyrazole derivative(Ia):compound(B-20:pyridate)=3:2~1:50
 pyrazole derivative(Ib):compound(B-1:atrazine)=2:1~1:50
 pyrazole derivative(Ib):compound(B-2:cyanazine)=2:1~1:50
 pyrazole derivative(Ib):compound(B-3:metribuzin)=3:1~1:25
 pyrazole derivative(Ib):compound(B-4:linuron)=2:1~1:50
 50 pyrazole derivative(Ib):compound(B-5:metbenzuron)=1:2~ 1:100
 pyrazole derivative(Ib):compound(B-6:bentazone)=6:1~1:3
 pyrazole derivative(Ib):compound(B-7:dicamba)=1:1~1:50
 pyrazole derivative(Ib):compound(B-8:chlorypyridid)=4:3~1:12
 pyrazole derivative(Ib):compound(B-9:2,4-D)=2:1~1:50
 55 pyrazole derivative(Ib):compound(B-10:bromoxynil)=1:1~1:50
 pyrazole derivative(Ib):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(Ib):compound(B-12:metolachlor)=2:1~ 1:25
 pyrazole derivative(Ib):compound(B-13:pendimethalin)=2:1~ 1:25
 pyrazole derivative(Ib):compound(B-14:nicosulfuron)=1:3~ 40:1

pyrazole derivative(lb):compound(B-15:rimsulfuron)=1:3~ 40:1
 pyrazole derivative(lb):compound(B-16:imazethapyr)=1:6~ 40:1
 pyrazole derivative(lb):compound(B-17:primisulfuron)=1:3~ 40:1
 pyrazole derivative(lb):compound(B-18:dimethenamid)=2:1~ 1:50
 5 pyrazole derivative(lb):compound(B-19:acetochlor)=2:1~1:50
 pyrazole derivative(lc):compound(B-1:atrazine)=2:1~1:50
 pyrazole derivative(lc):compound(B-2:cyanazine)=2:1~1:50
 pyrazole derivative(lc):compound(B-3:metribuzin)=3:1~1:25
 pyrazole derivative(lc):compound(B-4:linuron)=2:1~1:50
 10 pyrazole derivative(lc):compound(B-6:bentazone)=1:2~1:100
 pyrazole derivative(lc):compound(B-7:dicamba)=1:1~1:50
 pyrazole derivative(lc):compound(B-8:chlorypyralid)=4:3~1:12
 pyrazole derivative(lc):compound(B-9:2,4-D)=2:1~1:50
 pyrazole derivative(lc):compound(B-10:bromoxynil)=1:1~1:50
 15 pyrazole derivative(lc):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(lc):compound(B-12:metolachlor)=2:1~ 1:25
 pyrazole derivative(lc):compound(B-13:pendimethalin)=2:1~ 1:25
 pyrazole derivative(lc):compound(B-14:nicosulfuron)=1:3~ 40:1
 pyrazole derivative(lc):compound(B-15:rimsulfuron)=1:3~ 40:1
 20 pyrazole derivative(lc):compound(B-16:imazethapyr)=1:6~ 40:1
 pyrazole derivative(lc):compound(B-17:primisulfuron)=1:3~ 40:1
 pyrazole derivative(lc):compound(B-18:dimethenamid)=2:1~ 1:50
 pyrazole derivative(lc):compound(B-19:acetochlor)=2:1~1:50

25 The process for the production of the herbicide composition of the present invention will be explained hereinafter.

The herbicide composition of the present invention is obtained by mixing the pyrazole derivative of the above general formula (I) and at least one compound selected from a group of the above herbicide compounds with a liquid carrier such as a solvent or a solid carrier such as a mineral powder and forming the mixture into a preparation in the form of a wettable powder, an emulsifiable concentrate, a dust, granules, a flowable preparation or a solution. The preparation
 30 can be formed by adding surfactants such as an emulsifier, a dispersing agent, a spreading agent, a systemic agent and a stabilizer and other adjuvants as required.

When the herbicide composition of the present invention is used as a wettable powder, generally, a composition is prepared by 10 to 55 % by weight of the pyrazole derivative (I) and at least one selected from a group of the above herbicide compounds as active ingredients, 40 to 88 % by weight of a solid carrier and 2 to 5% by weight of a surfactant, and can be used as such. Further, when it is used in the forms of an emulsifiable concentrate and a flowable preparation, generally, it can be prepared by 5 to 50 % by weight of the pyrazole derivative (I) and at least one compound selected from a group of the above herbicide compounds as active ingredients, 35 to 90 % by weight of a solvent and 5 to 15 % by weight of a surfactant and other adjuvant.

On the other hand, when herbicide composition of the present invention is used in the form of a dust, generally, it
 40 can be prepared by mixing 1 to 15 % by weight of the pyrazole derivative (I) and at least one compound selected from a group of the above herbicide compounds as active ingredients and 85 to 99 % by weight of a solid carrier. Further, when it is used in the form of granules, it can be prepared by mixing 0.1 to 15 % by weight of the pyrazole derivative (I) and at least one compound selected from a group of the above herbicide compounds as active ingredients, 80 to 97.9 % by weight of a solid carrier and 2 to 5 % by weight of a surfactant. The above solid carrier can be selected from mineral fine powders, and the mineral fine powders include oxides such as diatomaceous earth and slaked lime, phosphates such as apatite, sulfates such as gypsum, and silicates such as talc, pyroferite, clay, kaolin, bentonite, acid clay, white carbon, powdered quartz and powdered silica.

The liquid carrier includes paraffin- or naphthene-based hydrocarbons such as kerosene, mineral oil and spindle oil, aromatic hydrocarbons such as benzene, toluene and xylene, chlorinated hydrocarbons such as o-chlorotoluene, trichloromethane and trichloroethylene, alcohols such as cyclohexanol, amyl alcohol and ethylene glycol, alcohol ethers such as ethylene glycol monomethyl ether and ethylene glycol monoethyl ether, ketones such as isophorone, cyclohexanone and cyclohexenyl-cyclohexanone, ethers such as butyl cellosolve, dimethyl ether and methyl ethyl ether, esters such as isopropyl acetate, benzyl acetate and methyl phthalate, amides such as dimethylformamide, nitriles such as acetonitrile and propionitrile, sulfoxides such as dimethylsulfoxide, organic solvents such as mixtures of these, and
 55 water.

Further, the surfactant can be selected from anionic ones (alkylbenzenesulfonate, alkylsulfonate and lauric amide sulfonate), nonionic ones (polyoxyethylene octyl ether, polyethylene glycol laurate and sorbitan alkyl ester), cationic ones (dimethylaurylbenzylammonium chloride, laurylamine and stearyltrimethylammonium chloride) and amphoteric ones (amino acid and betaine).

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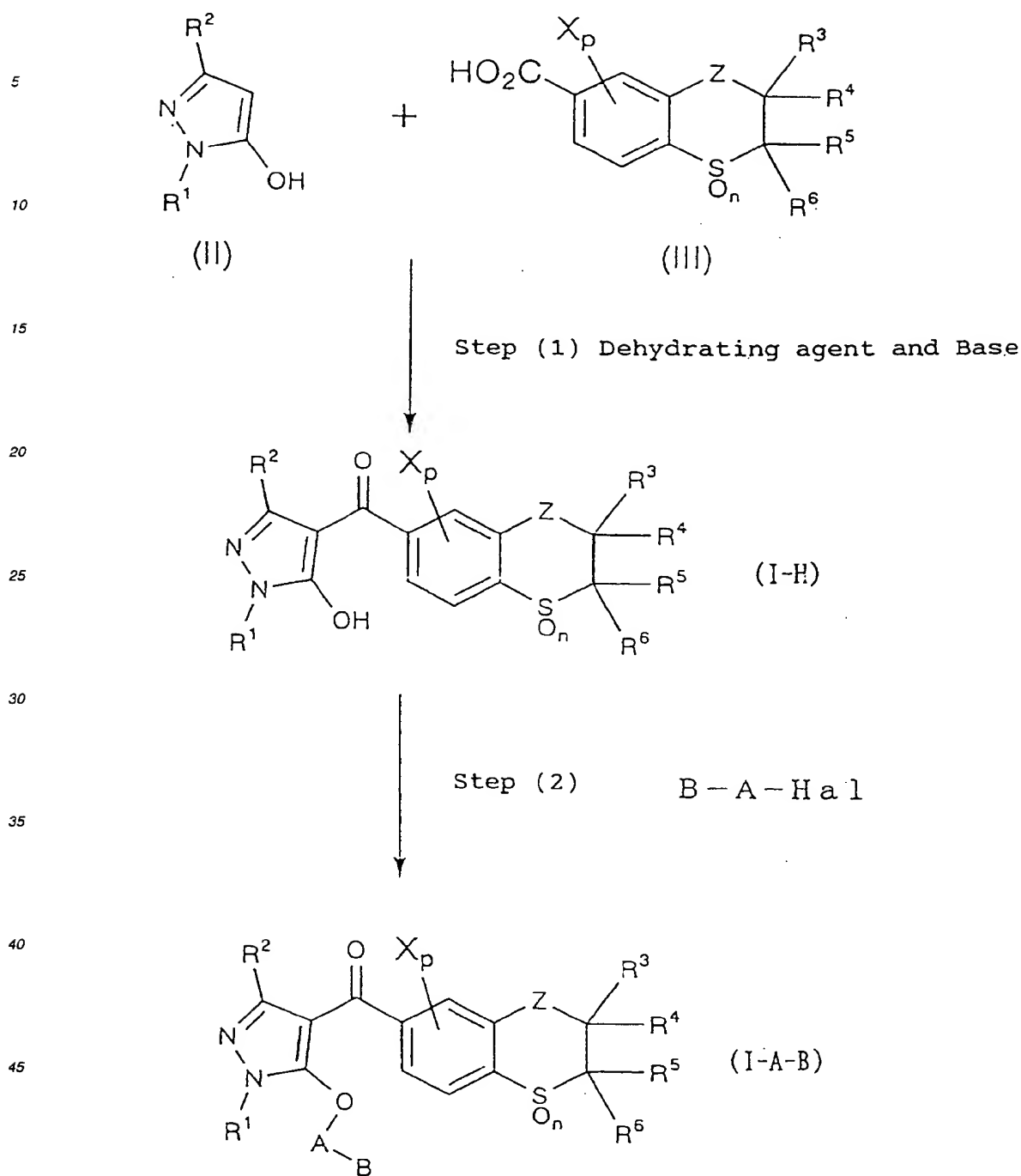
For improving the properties of the preparation and enhancing the herbicidal efficacy of the preparation, the herbicide composition of the present invention may contain polymer compounds or adjuvants such as sodium alginate, carboxymethyl cellulose, carboxyvinyl polymer, gum arabic and hydroxypropylmethylcellulose.

The herbicide composition of the present invention can simultaneously control gramineous weeds and broad-leaved weeds at a low dosage without damaging useful crops (i.e., without phytotoxicity) by soil treatment or foliar treatment for upland crops such as corn, Indian millet and the like before or after the germination of the weeds.

Further, the herbicide composition of the present invention exhibits excellent herbicidal efficacy against weeds in orchards and non-agricultural land (factory field, railroad site, roadside, river bed and fallow field) by soil treatment or foliar treatment.

The herbicide composition of the present invention is applied in an amount, as active ingredients, of approximately 10 to 1,000 g, preferably 70 to 700 g, per hectare. When it is applied to plant stalks and leaves, it is diluted to approximately 100 to 100,000 ppm, preferably 250 to 50,000 ppm, before use.

The pyrazole derivative of the general formula (I) can be produced by the following production process.



In the above reaction scheme, $R^1 \sim R^6$, X , p , n , A , B and Z are as defined in the general formula (I), and Hal is a halogen atom.

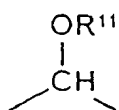
Some of thiochromancarboxylic acids of the general formula (III) used as a starting material for the production of the pyrazole derivative of the general formula (I) are already known, and the process for the production thereof is described in International Laid-open Patent Publications WO93/18031, WO94/01431 and WO95/04054. Those which are not known can be produced according to the process described in the above International Laid-open Patent Publications or U. S. Patent 5,035,793.

Further, the thiochromancarboxylic acid of the general formula (III) can be produced by one of the following produc-

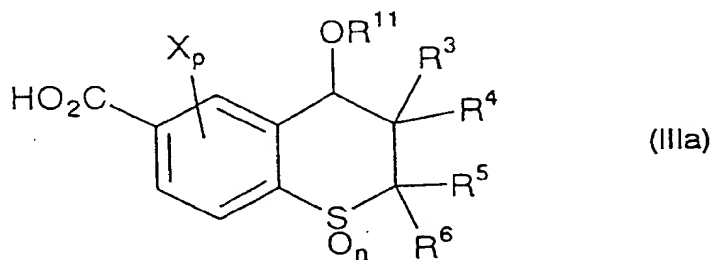
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tion schemes depending upon the structure of substituent. In the reaction schemes, R^1 to R^{12} , X, p, n, A, B and Z are as defined in the general formula (I), Hal is a halogen atom, and q is 1 or 2.

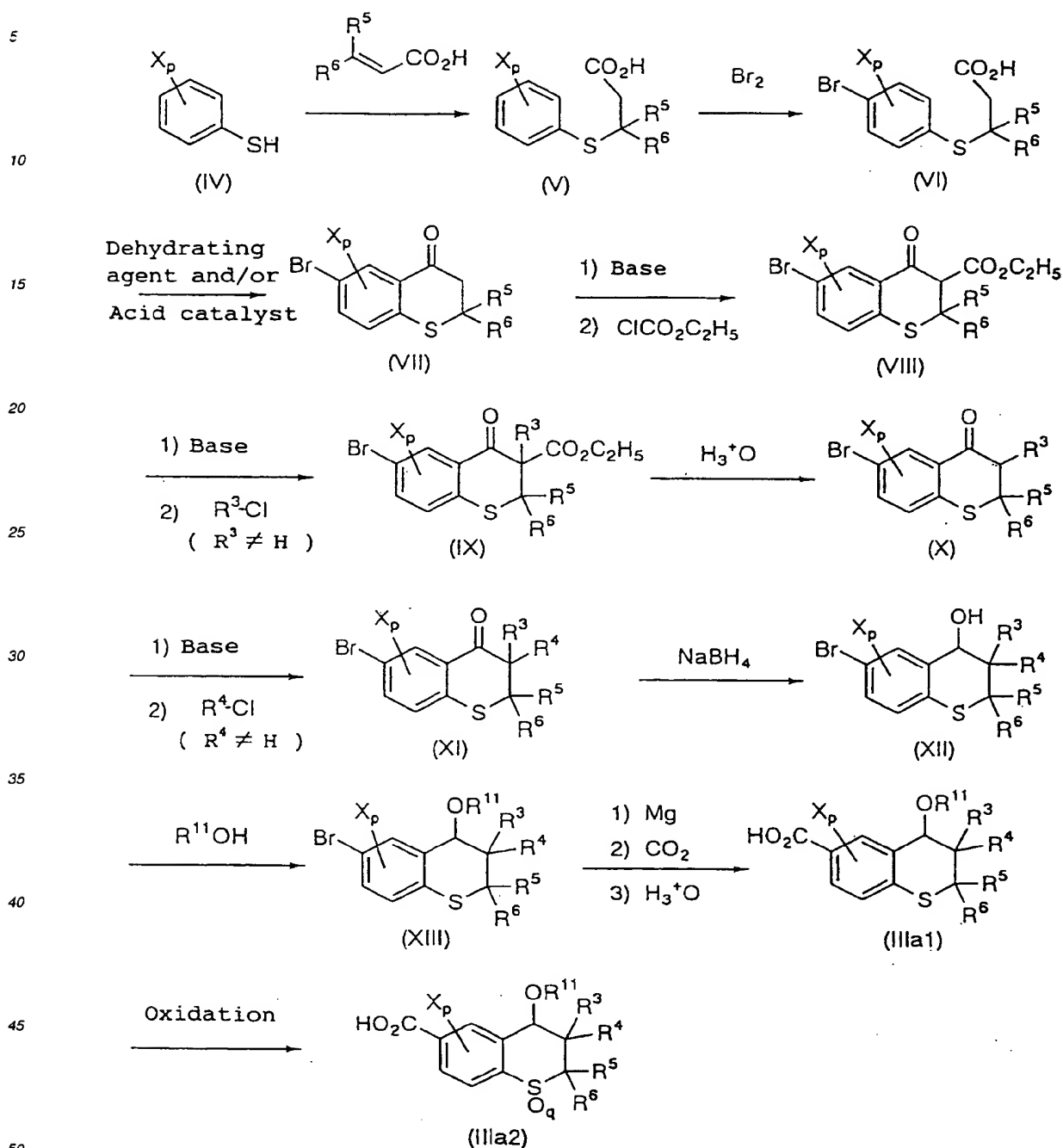
The thiochromancarboxylic acid of the general formula (III) in which Z is



i.e., a thiochromancarboxylic acid of the following general formula (IIIa), can be produced by the method in any one of the production schemes 1 ~ 3.



Production scheme 1



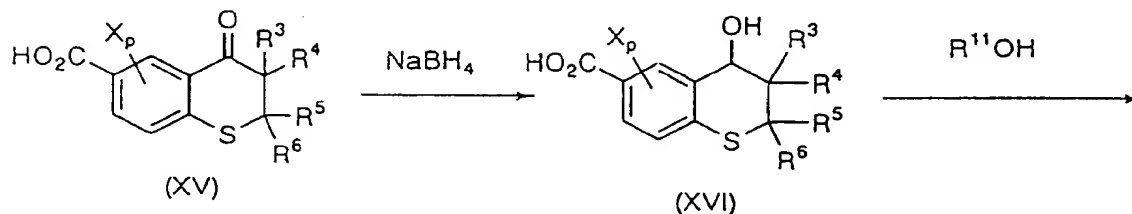
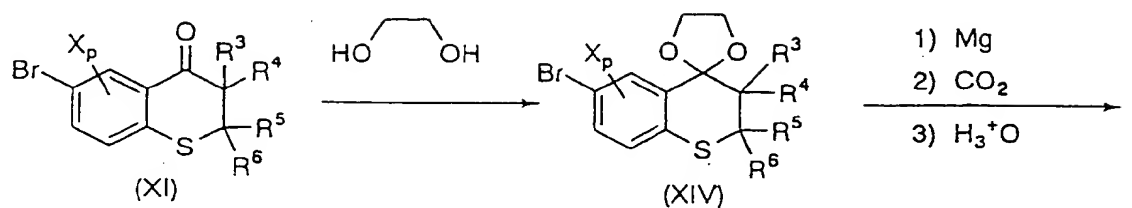
In the production scheme 1, a compound (IIIa1) is a thiochromancarboxylic acid of the general formula (IIIa) in which

$n = 0$.

A compound (IIIa2) is a thiochromancarboxylic acid of the general formula (IIIa) in which

$n = 1$ or 2 .

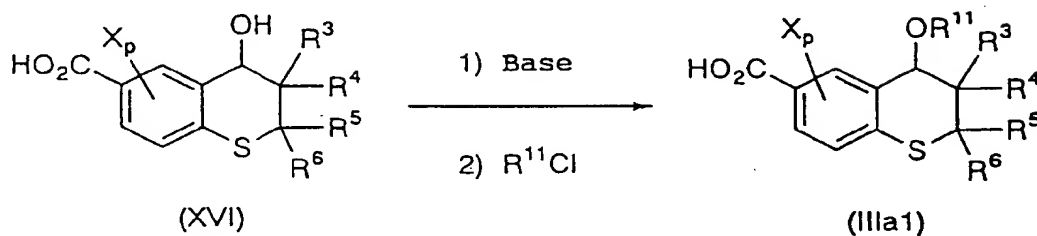
Production scheme 2



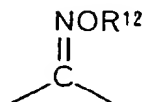
(IIIa1)

The following is
the same as in Scheme 1.

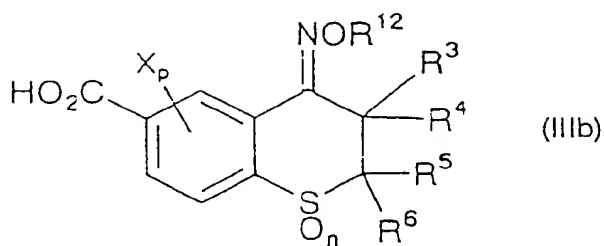
Production scheme 3



To be followed in the same manner as in Scheme 1.
The compound of the general formula (III) in which Z is

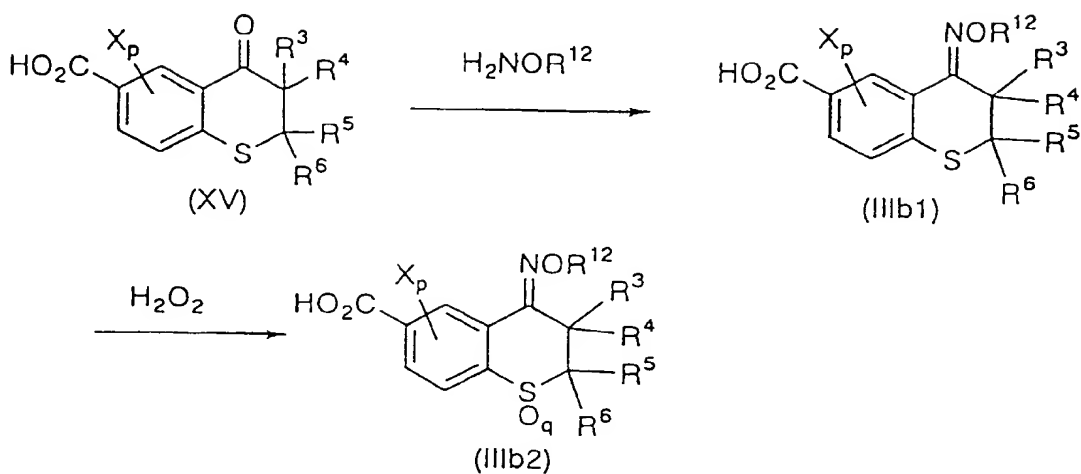


i.e., a thiochromancarboxylic acid of the following general formula (IIIb), can be produced by the method in any one of the production schemes 4 ~ 5.



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Production scheme 4

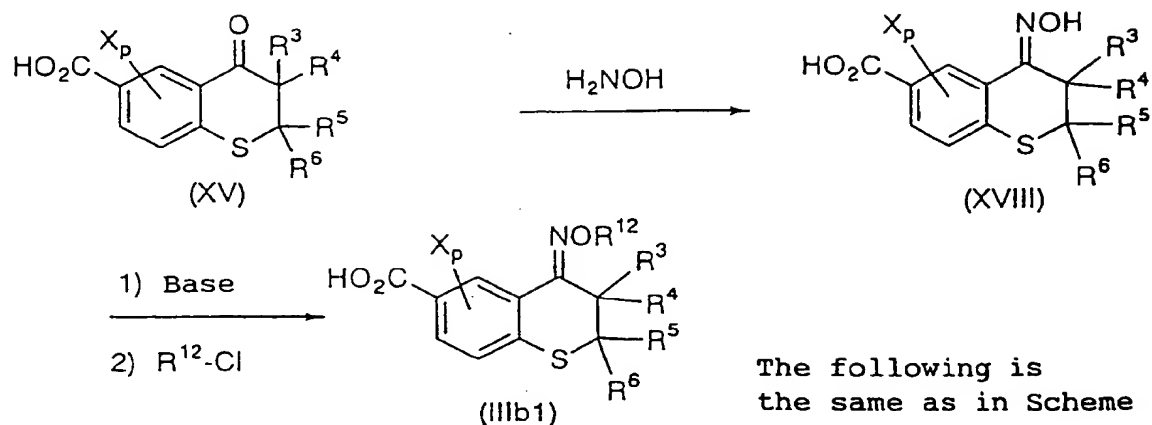


In the production scheme 4, a compound (IIIb1) is a thiochromancarboxylic acid of the general formula (IIIb) in which

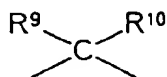
$n = 0$.

A compound (IIIb2) is a thiochromancarboxylic acid of the general formula (IIIb) in which

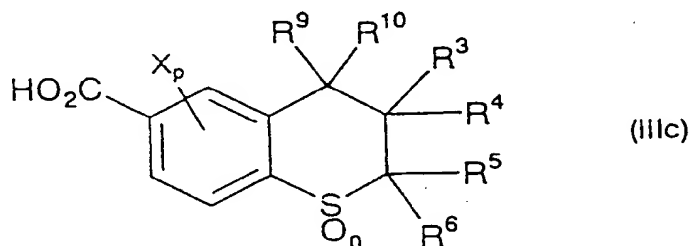
$n = 1$ or 2 .

Production scheme 5

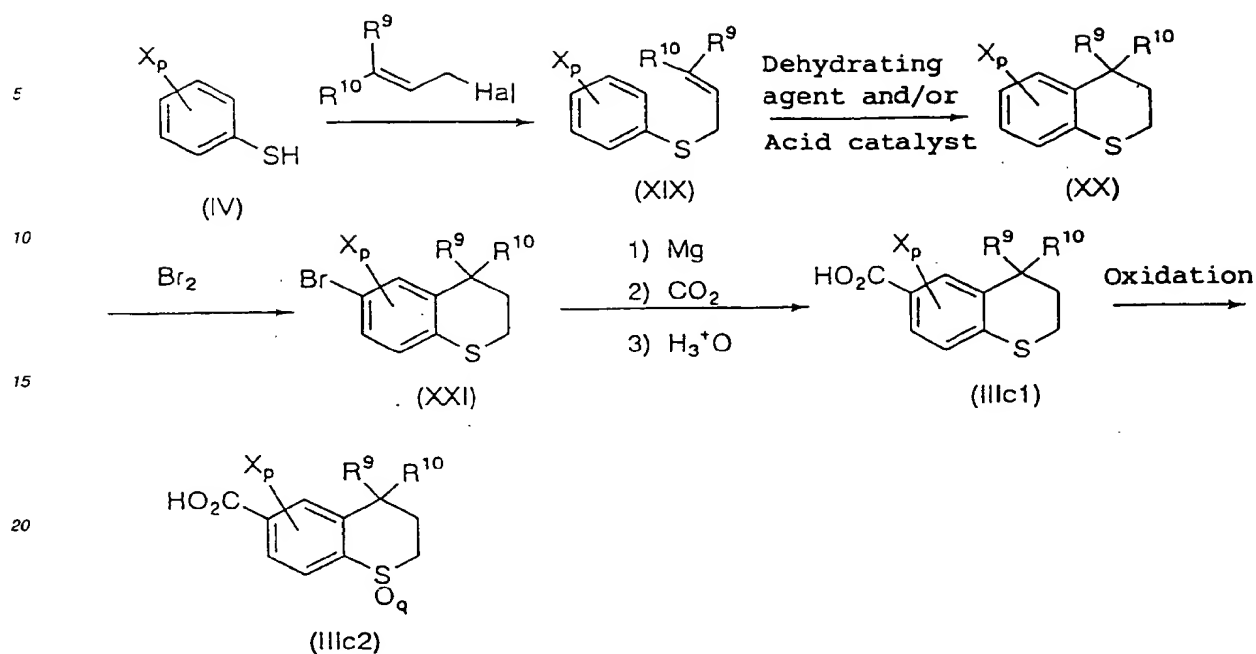
The thiochromancarboxylic acid of the general formula (III) in which Z is



i.e., a thiochromancarboxylic acid of the following general formula (IIIc), can be produced by the method in any one of the production schemes 6 ~ 12.

Production scheme 6

The production scheme 6 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which R³, R⁴, R⁵ and R⁶ are all hydrogen atoms.



In the production scheme 6, a compound (IIIc1) is a thiochromancarboxylic acid of the general formula (IIIc) in which

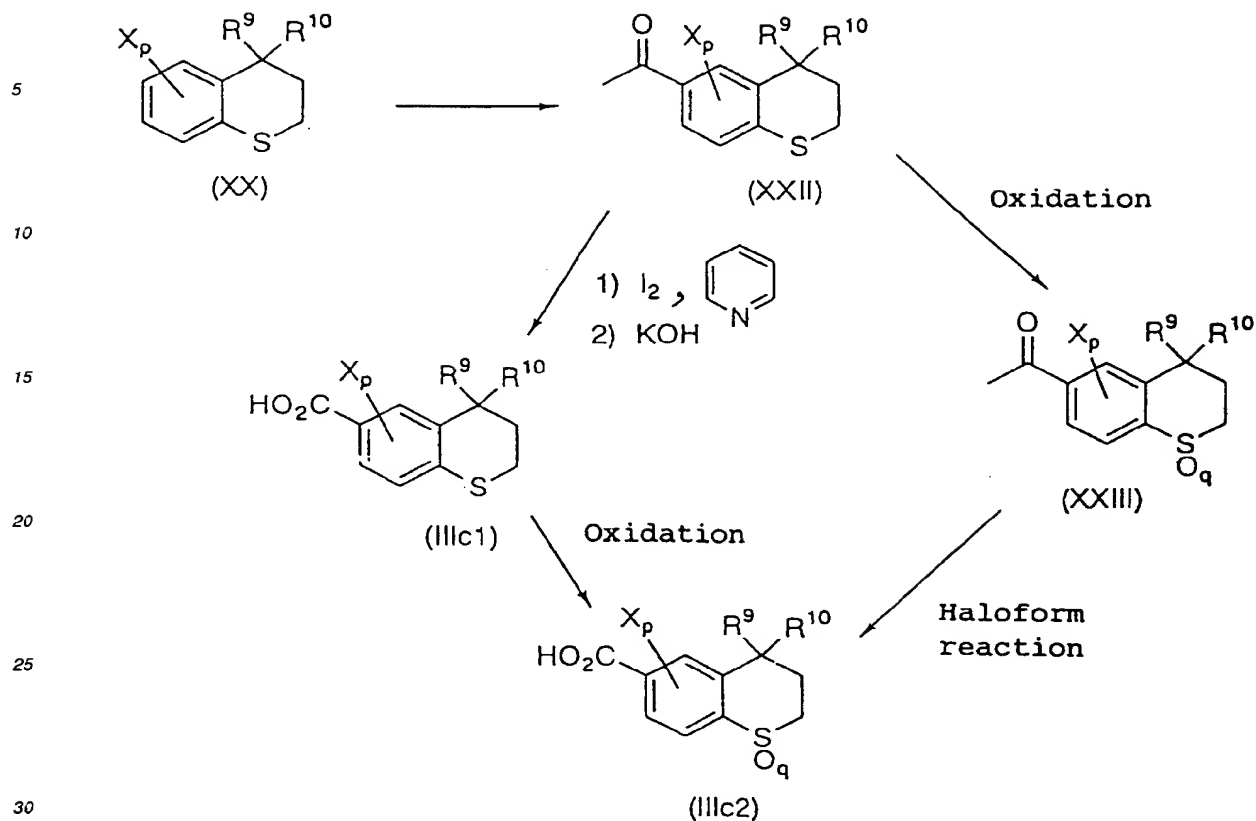
$R^3 = R^4 = R^5 = R^6 = \text{hydrogen atom}$, and $n = 0$ (sulfide).

A compound (IIIc2) is a thiochromancarboxylic acid of the general formula (IIIc) in which

$R^3 = R^4 = R^5 = R^6 = \text{hydrogen atom}$, and $n = 1$ (sulfoxide) or 2 (sulfone).

Production scheme 7

The thiochromancarboxylic acid of the general formula (IIIc1) or (IIIc2) in the above production scheme 7 can be also produced by the production scheme 7.

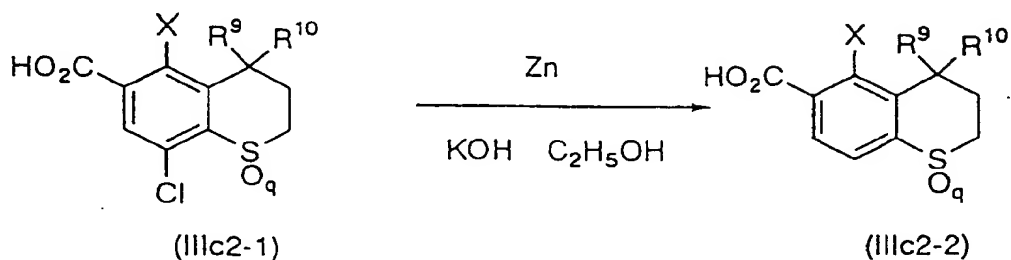
**Production scheme 8**

A thiochromancarboxylic acid of the general formula (IIIc2) in which

$p = 1$ and

Position of substituent X = 5-position on the thiochroman ring,

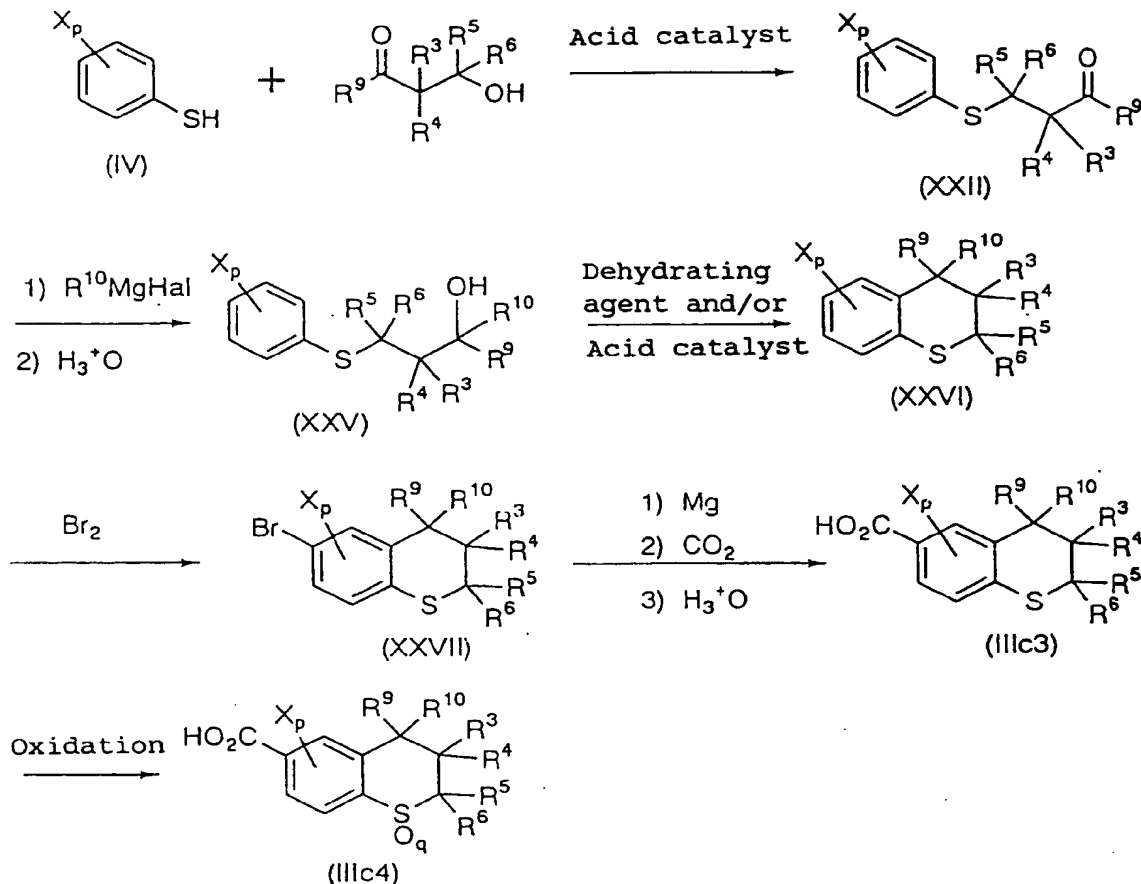
can be also produced by the production scheme 7.

**Production scheme 9**

The production scheme 9 is directed to a process for the production of a thiochromancarboxylic acid of the general

formula (IIIc) in which

R^{10} = C_1 ~ C_4 alkyl group or C_2 ~ C_5 alkenyl group and
 R^9 = C_1 ~ C_4 alkyl group or C_1 ~ C_4 haloalkyl group.



In the production scheme 9, a compound (IIIc3) is a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = C_1 ~ C_4 alkyl group or C_2 ~ C_5 alkenyl group,
 R^9 = C_1 ~ C_4 alkyl group or C_1 ~ C_4 haloalkyl group, and
 $n = 0$.

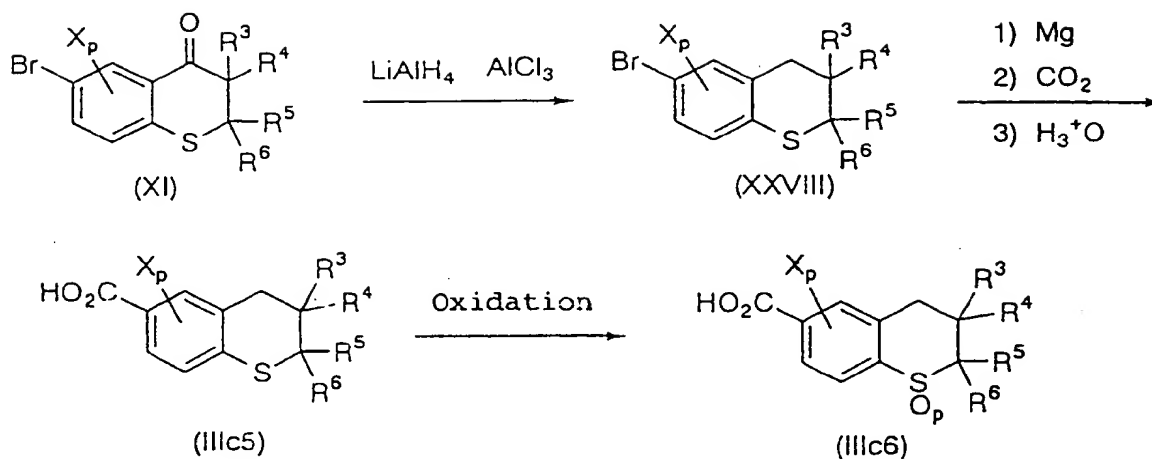
A compound (IIIc4) is a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = C_1 ~ C_4 alkyl group or C_2 ~ C_5 alkenyl group,
 R^9 = C_1 ~ C_4 alkyl group or C_1 ~ C_4 haloalkyl group, and
 $n = 1$ or 2 .

Production scheme 10

The production scheme 10 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which

$R^9 = R^{10} =$ hydrogen atom.



In the production scheme 10, a compound (IIIc5) is a compound of the general formula (IIIc) in which

$R^9 = R^{10} = \text{hydrogen atom}$, and
 $n = 0$.

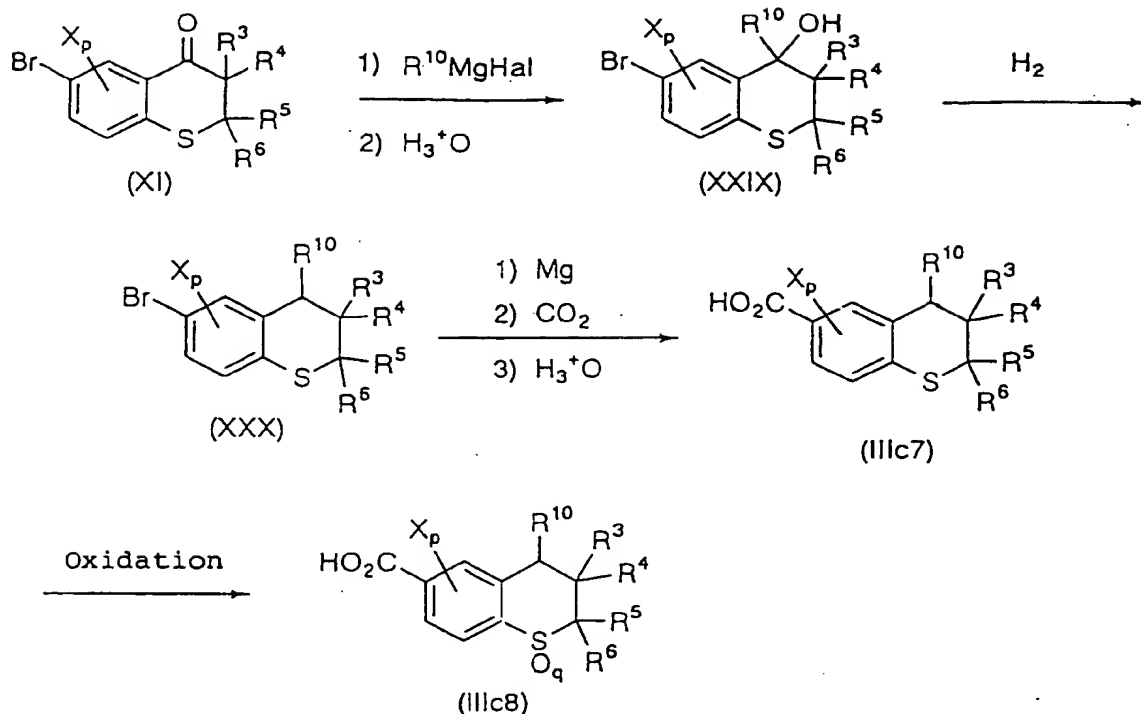
A compound (IIIc6) is a compound of the general formula (IIIc) in which

$R^9 = R^{10} = \text{hydrogen atom}$, and
 $n = 1 \text{ or } 2$.

Production scheme 11

The production scheme 11 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which

$R^9 = \text{hydrogen atom}$.



In the production scheme 11, a compound (IIIc7) is a compound of the general formula (IIIc) in which

R^9 = hydrogen atom, and
 $n = 0$.

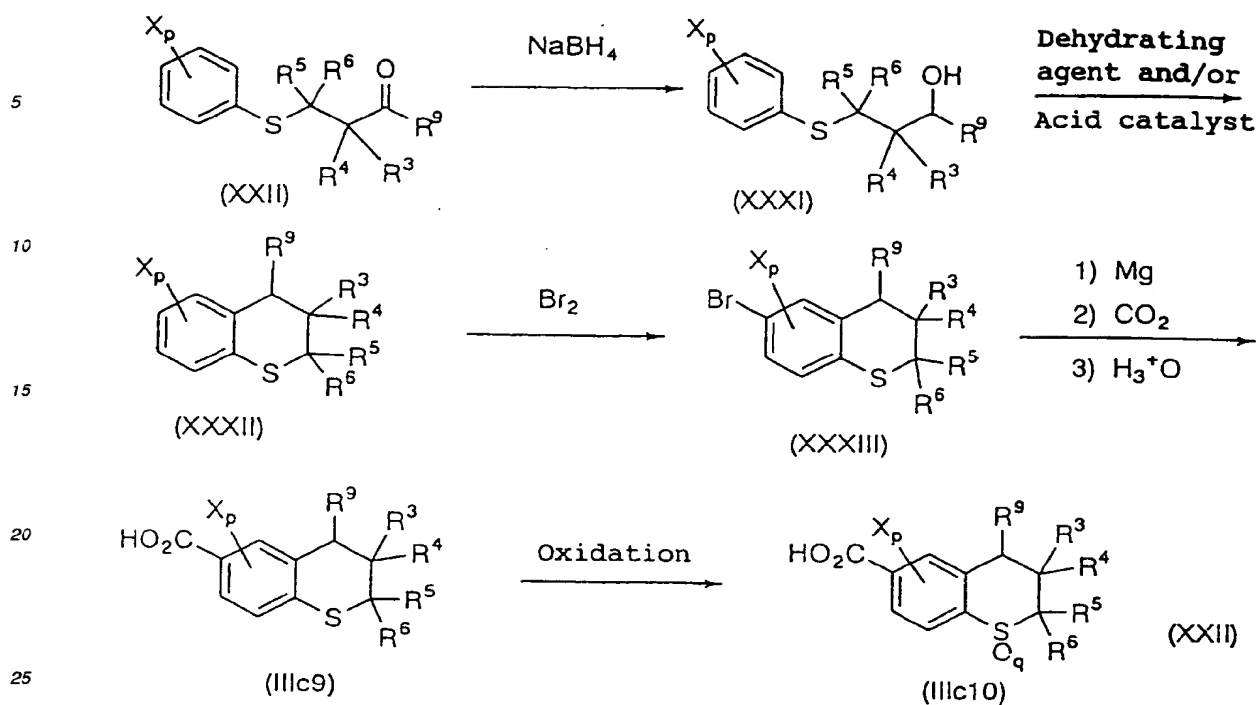
A compound (IIIc8) is a compound of the general formula (IIIc) in which

R^9 = hydrogen atom, and
 $n = 1$ or 2 .

Production scheme 12

The production scheme 12 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = hydrogen atom.



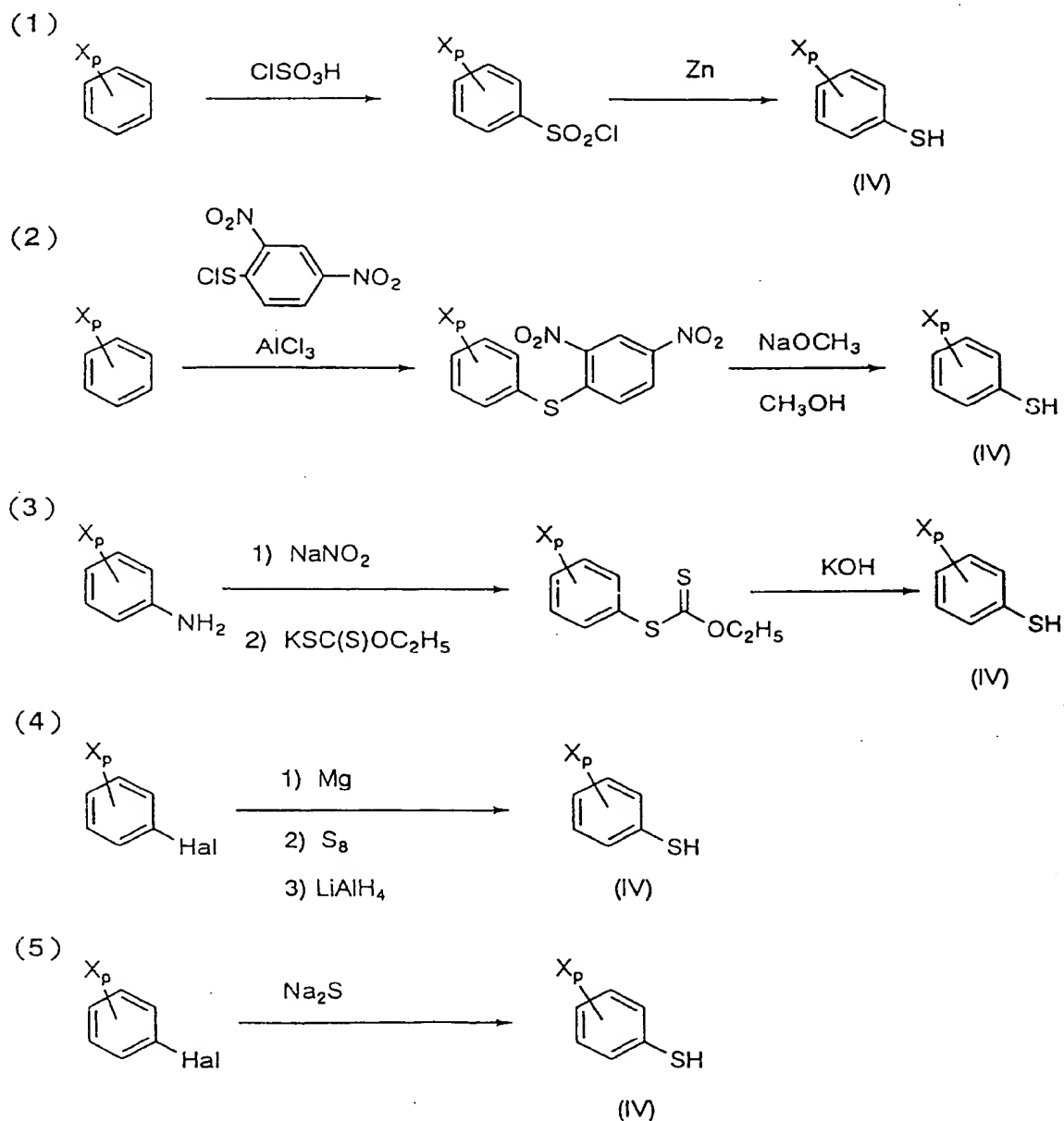
In the production scheme 12, a compound (IIlc9) is a thiochromancarboxylic acid of the general formula (IIlc) in which

R^{10} = hydrogen atom, and
 $n = 0$.

A compound (IIlc10) is a compound of the general formula (IIlc) in which

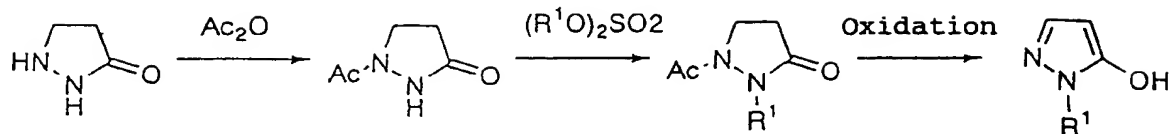
R^{10} = hydrogen atom, and
 $n = 1$ or 2 .

Triphenols of the general formula (IV) (production scheme 1), as a starting material for the production of the thiochromancarboxylic acid, can be produced by one of the following methods depending upon their substituents. In the following reaction scheme, X and p are as defined in the general formula (I) and Hal is a halogen atom.

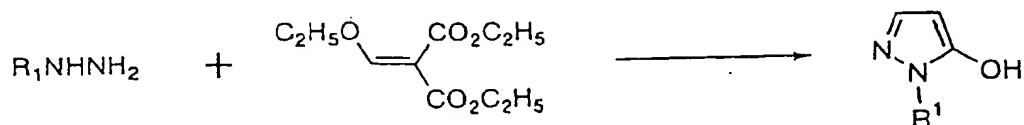


5-Hydroxypyrazoles of the general formula (II), as a starting material for the production of the pyrazole derivative (I) of the present invention, can be produced by one of the following methods depending upon their substituents. In the following reaction schemes, R^1 and R^2 are as defined in the general formula (I).

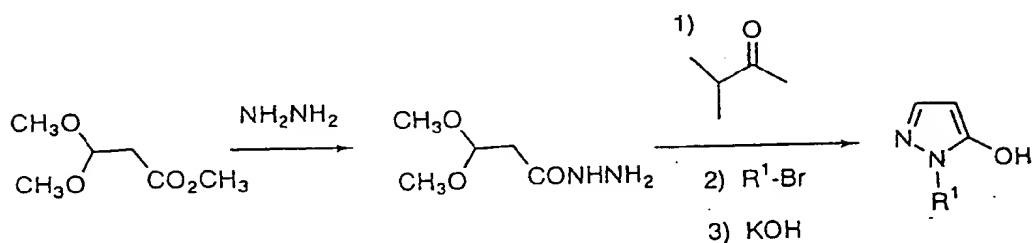
(1) Process mentioned in East German Pat. No. 83145



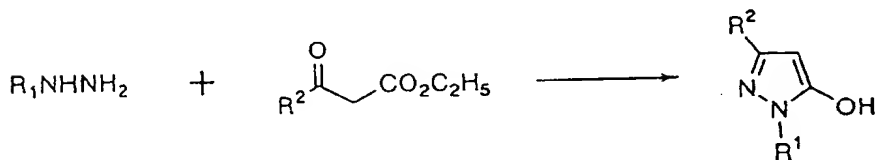
10 (2) Process mentioned in US Pat. No. 4744815



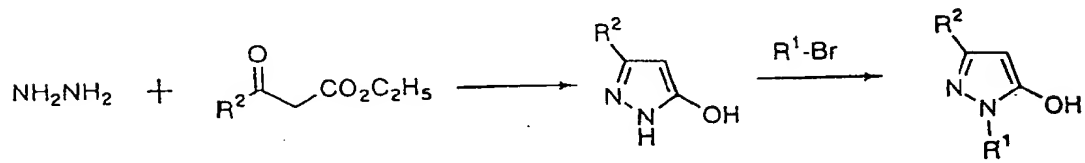
20 (3) Process mentioned in Japanese Laid-open No. Hei-3-44375



35 (4)



45 (5)



55 The above (1)~(3) are directed to a process for the production of 5-hydroxypyrazoles of the general formula (II) in which

R^2 = hydrogen atom.

60 The above (5) ~ (6) are directed to a process for the production of 5-hydroxypyrazoles of the general formula (II) in which

R^2 = C_1 ~ C_4 alkyl group, C_1 ~ C_4 haloalkyl group or C_2 ~ C_4 alkoxy group.

Examples

The present invention will be specifically explained with reference to Production Referential Examples, Preparation Examples and Examples hereinafter.

Production Referential Examples

Production Referential Example 1

Synthesis of 4-methoxy-5-methyl-6-(5-cyclohexylcarbonyloxy-1-ethylpyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ia-7)

As a starting material, 4-methoxy-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide corresponding to pyrazole derivative (I-H) was used. 0.4 Gram (1.1 mmol) thereof was dissolved in 4 ml of methylene chloride, and 0.22 g (2.2 mmol) of triethylamine as a base and 0.19 g (1.3 mmol) of cyclohexylcarbonyl chloride corresponding to compound B-A-Hal as a reaction reagent were added. The mixture was allowed to react at room temperature for 8 hours. A saturated sodium carbonate aqueous solution was added to the reaction mixture, and the resultant mixture was extracted with ethyl acetate. An organic layer was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the resultant oil was purified by flush column chromatography (Wako Gel C-300; hexane/ethyl acetate = 1:1) to give 0.28 g (yield 54 %) of 4-methoxy-5-methyl-6-(5-cyclohexylcarbonyloxy-1-ethylpyrazol-4-yl)carbonylthiochroman-1,1-dioxide.

Referential Production Example 2

Synthesis of 4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-1)

A 100-ml eggplant type flask was charged with 1.1 g (2.9 mmol) of 4-methoxyimino-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide corresponding to pyrazole derivative (I-H) as a starting material, and 20 ml of methylene chloride was added and allowed to dissolve it. Then, a solution of 0.41 g of potassium carbonate in 20 ml of distilled water was added. Further, a solution of 0.6 g (4.2 mmol) of n-propanesulfonyl chloride corresponding to compound B-A-Hal as a reaction reagent in 5 ml of methylene chloride was added, and further, 0.05 g of benzyltriethylammonium chloride as a catalyst was added. The mixture was allowed to react at room temperature for 24 hours with stirring. After the reaction was completed, a methylene chloride layer was separated and dried over anhydrous sodium sulfate, and then the methylene chloride was distilled off under reduced pressure. The resultant oily substance was purified with a column packed with silica gel. A mixture of ethyl acetate with n-hexane was used as a developer solution.

By the above procedures, 0.88 g of 4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-1) was obtained as a solid. The yield thereof was 62 %.

Referential Production Example 3

Synthesis of 4-methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-2)

0.4 Gram (1.1 mmol) of 4-methoxyimino-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide corresponding to pyrazole derivative (I-H), 0.23 g (1.2 mmol) of phenacyl bromide corresponding to compound B-A-Hal and 0.15 g of potassium carbonate were added to 10 ml of acetone, and the mixture was stirred under heat for 8 hours. Insolubles were removed by filtration, and then the acetone was distilled off. The residue was dissolved in ethyl acetate, and the mixture was washed with a saturated sodium chloride aqueous solution and dried over sodium sulfate. The ethyl acetate was distilled off under reduced pressure, and the residue was subjected to column chromatography (hexane/ethyl acetate) to give 4-methoxyimino-5-methyl-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-2) at a yield of 52 %.

Referential Production Example 4

Synthesis of 4-methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-3)

4-Methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Com-

pound Ib-3) was obtained in the same manner as in Referential Production Example 2 except that the reaction reagent was replaced with p-toluenesulfonyl chloride corresponding to compound B-A-Hal.

Referential Production Example 5

Synthesis of 4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound 1c-1)

7.4 Grams (0.026 mol) of 4,4,5,8-tetramethylthiochroman-6-carboxylic acid-1,1-dioxide corresponding to thiochroman carboxylic acid (IIIc), 3.4 g (0.03 mol) of 1-ethyl-5-hydroxypyrazole corresponding to 5-hydroxypyrazole (II) and 6.22 g (0.03 mol) of DCC (N,N'-dicyclohexylcarbodiimide) were all together added to 50 ml of tert-amyl alcohol, and the mixture was stirred at room temperature for 30 minutes. Then, 1.8 g (0.013 mol) of anhydrous potassium carbonate was added. The reaction mixture was allowed to react at 80°C for 8 hours, and the reaction solvent was distilled off under reduced pressure. The resultant residue was dispersed in a 5 % potassium carbonate aqueous solution and ethyl acetate to separate it into two layers. Further, the aqueous layer was adjusted to a pH of 1 with 5 % hydrochloric acid, and the formed solid was recovered by filtration to give 6.13 g (yield 62 %) of 4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound 1c-1).

Referential Production Example 6

Synthesis of 4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound 1c-2)

0.7 Gram (1.9 mmol) of the 4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound 1c-1) corresponding to pyrazole derivative (I-H), obtained in Referential Production Example 5, was dissolved in 8 ml of methylene chloride. Then, a solution of 0.51 g (3.8 mmol) of potassium carbonate in 5 ml of water was added, and further, 0.49 g (3.8 mmol) of ethanesulfonyl chloride and 0.05 g (0.2 mmol) of benzyltriethylammonium chloride, corresponding to compound B-A-Hal, were added. The mixture was allowed to react at room temperature for 2 hours, and further refluxed under heat for 2 hours. The reaction mixture was allowed to cool, and then methylene chloride layer was recovered and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the resultant oil was purified by silica gel column chromatography to give 0.73 g (yield 82 %) of 4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound 1c-2).

Table 1 shows physical property data of the compounds obtained in the above Referential Production Examples 1 ~ 6.

Table 1

Prepn Ex. No.	Compd No.	N.M.R.(ppm) Internal standard: tetramethylsilane Solvent: deu- terochloroform	I.R. (cm ⁻¹) KBr tablet	mp (°C)
1	la-7	1.42(3H,t,J=7.3Hz) 1.3- 2.0(10H,m) 2.33(3H,s)2.3- 2.8(3H,m) 3.1- 3.3(1H,m)3.47(3H,s) 3.6- 3.8(1H,m) 3.99(2H,q,J=7.3Hz) 4.52(1H,t,J=2.9Hz) 7.44(1H,d,J=8.2Hz)7.61 (1H,s) 7.86(1H,d,J=8.2Hz)	2970,1800, 1670,1300, 1140	139.0-141.0
2	lb-1	1.18(3H,t)1.52(3H,t)2.00 2.20(2H,m) 2.52(3H,s)3.35 (4H,t)3.73(2H,t 4.06(3H,s)4.23(2H,q) 7.45(H,s 7.48(H,d)7.96(H,d)	3000,2960, 1665, 1135,1325, 1190,1395	146.0-150.7
3	lb-2	1.51(3H,t)2.41(3H,s) 3.20- 3.40(4H,m) 4.02(3H,s)4.28(2H,q) 6.19(2H,s) 7.19(H,s)7.30-8.10(7H,m)	2950,1710, 1650,1320, 1130	-----
4	lb-3	1.49(3H,t)2.47(3H,s) 2.49(3H,s 3.3-3.5(4H,m)4.05 (3H,s)4.17(2H,q) 7.35(1H,s)7.4- 8.0(6H,m)	2950,1680, 1320,1130	glass -like
5	lc-1	1.45(3H,t)1.55(6H,S) 2.30- 2.50(2H,m) 2.50(3H,s)2.80(3H,s) 3.40- 3.60(2H,m) 4.10(2H,q)6.20(H,s) 7.20(H,s)	2550-3500, 2950,3000, 1630,1290, 1130	208.8-209.3
6	lc-2	1.50(3H,t)1.60(6H,s) 1.70(3H,t) 2.30-2.60(2H,m)2.50(3H,s) 2.80(3H,s) 3.30- 3.60(2H,m)3.80(2H,q) 4.20(2H,q) 7.10(H,s)7.40(H,s)	2940,3000, 1660,1180, 1140,1290, 1380	164.1-165.7

Preparation Examples

The method of forming preparations will be explained with reference to Preparation Examples hereinafter. "Part" in the following Preparation Examples stands for part by weight.

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Preparation Example 1 [Wettable powder]

Compound (Ia-1), (Ib-1) or (Ic-1)	5 parts
Compound (B-1)	25 parts
Diatomaceous earth	52 parts
White carbon	15 parts
Sodium alkylbenzenesulfonate	2 parts
Sodium ligninsulfonate	1 part

The above components were mixed and homogeneously mixed and pulverized to give 100 part of a wettable powder.

Preparation Example 2 [Emulsifiable concentrate]

Compound (Ia-1), (Ib-1) or (Ic-1)	5 parts
Compound (B-2)	25 parts
Xylene	30 parts
Methylnaphthalene	20 parts
Sorpol 2680 (Surfactant supplied by Toho Chemical Co., Ltd.)	20 parts

The above components were homogeneously dissolved and mixed to give 100 parts of an emulsifiable concentrate.

Preparation Example 3 [Dust]

Compound (Ia-1), (Ib-1) or (Ic-1)	0.3 part
Compound (B-3)	1.7 part
Diatomaceous earth	20 parts
Talc	78 parts

The above components were mixed and homogeneously mixed and pulverized to give 100 parts of a dust.

Preparation Example 4 [Flowable preparation]

Compound (Ia-1), (Ib-1) or (Ic-1)	4 parts
Compound (B-4)	25 parts
Methyl cellulose	0.3 part
Colloidal silica	1.5 part
Sodium ligninsulfonate	1 part
Polyoxyethylene nonylphenyl ether	2 parts
Water	66.2 parts

The above components were fully mixed and dispersed, and the resultant mixture in a slurry state was wet-pulverized to give 100 parts of a stable flowable preparation.

Preparation Example 5 [Wettable powder]

97 Parts of clay (trade name: Zeaklite, supplied by Zeaklite Industry Co., Ltd.) as a carrier, 1.5 parts of alkylarylsulfonate (trade name: Neoplex, supplied by Kao-Atlas K.K.) as a surfactant and 1.5 parts by weight of a nonionic and anionic surfactant (trade name: Sorpol 800A, supplied by Toho Chemical Co., Ltd.) were homogeneously pulverized and mixed to prepare 90 parts of a carrier for a wettable powder. 10 Parts of Compound (Ia-1) ~ (Ia-5), (Ia-7), (Ib-1) ~ (Ib-3) or (Ic-1) or 10 parts of one of Compounds (B-1) ~ (B-2) was homogeneously mixed with, and pulverized together with, the above carrier to give wettable powders. A wettable powder containing one of (Ia-1) ~ (Ia-5), (Ia-7), (Ib-1) ~ (Ib-3) and (Ic-1) and a wettable powder containing one of Compound (B-1) ~ (B-20) were mixed in a predetermined mixing ratio (active ingredient ratio) to give wettable powders.

A biological test of the herbicide composition of the present invention will be shown as Examples hereinafter.

Example 1 [Foliar treatment test]

Seeds of weeds such as cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass and seeds of corn were sown in 1/2,000-are Wagner pots filled with upland soil, and covered with upland soil. Then, the seed were grown in a greenhouse, and when these weeds were at a 1.5 ~ 2.5 leaves stage, a predetermined amount of the herbicide obtained in the above Preparation Example 5 was suspended in water and uniformly sprayed onto foliar portions in a solution amount of 1,000 liters/hectare. Thereafter, the weeds were grown in a greenhouse, and 20 days after the treatment, the herbicide was determined for phytotoxicity to crops and herbicidal efficacy according to the following standard.

The herbicidal efficacy (weed control ratio %) was determined by measuring an above-ground green forage weight in a chemical-treated plot and an above-ground green forage weight in a non-treated plot and applying the measurement values to the following equation (A).

$$\text{Weed control ratio (\%)} = (1 - \frac{\text{above-ground green forage weight in treated plot}}{\text{above-ground green forage weight in non-treated plot}}) \times 100$$

The phytotoxicity was evaluated on the basis of the following six ratings.

Degrees of phytotoxicity

- 0 - No phytotoxicity to crops is observed.
- 1 - There is almost no phytotoxicity to crops.
- 2 - Phytotoxicity to crops is observed to some extent.
- 3 - Phytotoxicity to crops is observed.
- 4 - Phytotoxicity to crops is markedly observed.
- 5 - Crops almost died.

In a foliar treatment test, a herbicide is applied by spraying, and it is therefore difficult to bring the amount of the

herbicide to test weeds and crop into a completely constant value. Further, in both a foliar treatment test and a soil treatment test, it is also difficult to constantly bring test conditions such as temperature, humidity, hours of sunlight or water content in soil into completely constant values in the tests even if the tests are carried out in a greenhouse.

Therefore, the herbicidal efficacy of the single active ingredient of each of Compounds (B-1) ~ (B-20) varied in each test. However, the herbicidal efficacy (synergistic effect) of each herbicide composition of the present invention was evaluated on the basis of the results of the tests on single active ingredients carried concurrently, and was reliable.

Table 2 shows the foliar treatment test results of single active ingredients of Compounds (Ia-1), (Ia-2), (Ia-3), (Ia-4), (Ia-5) and (Ia-7) and Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17).

Tables 3 ~ 8 show the foliar treatment test results of herbicide compositions of Compounds (Ia-1), (Ia-2), (Ia-3), (Ia-4), (Ia-5) and (Ia-7) and Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17).

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Table 2

Foliar treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ia-1)	40	80	20	50	60	80	60	0
	20	80	20	30	40	60	30	0
(Ia-2)	40	50	50	30	40	60	50	0
	20	30	50	10	20	40	30	0
(Ia-3)	40	50	60	30	50	60	40	0
	20	30	40	10	30	40	30	0
(Ia-4)	40	50	50	30	40	50	40	0
	20	40	30	10	20	30	20	0
(Ia-5)	40	60	50	30	40	50	30	0
	20	40	30	10	20	30	20	0
(Ia-7)	40	40	50	40	50	50	40	0
	20	30	30	10	20	20	20	0
(B-1)	250	0	0	60	0	0	0	0
(B-10)	250	90	40	30	0	0	0	0
(B-7)	250	80	40	40	0	0	0	0
(B-6)	250	90	0	0	0	0	0	0
(B-20)	250	0	10	0	0	0	0	0
(B-3)	62	90	90	60	0	40	0	0
(B-2)	125	80	0	0	40	60	0	0
(B-4)	125	20	0	0	20	60	0	0
(B-9)	250	100	20	20	0	0	0	0
(B-14)	16	20	20	20	60	50	60	0
(B-15)	10	20	20	20	50	50	50	0
(B-16)	36	0	0	20	80	70	80	0
(B-17)	16	30	20	20	20	20	10	0
(a): Cocklebur (b): Velvet leaf (c): Slender amaranth (d): Green foxtail (e): Crabgrass (f): Barnyard grass								

Table 3 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-1)+(8-1) 40 + 250	100: 80: 20	90: 20: 70	100: 80: 20	80: 60: 20	100: 80: 20	100: 60: 40	0
(1a-1)+(8-10) 20 + 250	100: 98: 2	100: 52: 48	80: 51: 29	80: 40: 40	90: 60: 30	80: 30: 50	0
(1a-1)+(8-7) 40 + 250	100: 96: 4	70: 52: 18	100: 70: 30	90: 60: 30	100: 80: 20	80: 60: 20	0
(1a-1)+(8-6) 40 + 250	100: 98: 2	80: 20: 60	100: 50: 50	80: 60: 20	100: 80: 20	90: 60: 30	0
(1a-1)+(8-20) 40 + 250	100: 80: 20	90: 20: 70	60: 50: 10	80: 60: 20	100: 80: 20	90: 60: 30	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)
A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 4 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-2)+(B-1) 20 + 250	100 30 70	100 50 50	90 64 26	90 20 70	100 40 60	90 30 60	0
(1a-2)+(B-10) 40 + 250	100 95 5	100 70 30	70 51 19	90 40 50	80 60 20	80 50 30	0
(1a-2)+(B-7) 40 + 250	100 90 10	100 70 30	80 58 22	50 40 10	70 60 10	70 50 20	0
(1a-2)+(B-6) 40 + 250	100 95 5	100 50 50	90 30 60	80 40 40	100 60 40	90 50 40	0
(1a-2)+(B-20) 40 + 250	50 50 0	60 55 5	50 30 20	50 40 10	60 60 0	60 50 10	0
(1a-2)+(B-3) 40 + 62	100 95 5	100 95 5	100 72 28	90 40 50	100 76 24	100 50 50	0
(1a-2)+(B-2) 20 + 125	100 86 14	80 50 30	90 10 80	90 52 38	100 76 24	80 30 50	0
(1a-2)+(B-4) 40 + 125	100 60 40	100 50 50	90 30 60	90 52 38	100 84 16	100 50 50	0
(1a-2)+(B-9) 40 + 250	100 100 0	100 60 40	80 44 36	50 40 10	70 60 10	50 50 0	0
(1a-2)+(B-14) 40 + 16	80 60 20	60 60 0	50 44 6	80 76 4	80 80 0	80 80 0	0
(1a-2)+(B-15) 40 + 10	70 60 10	60 60 0	50 44 6	80 70 10	80 80 0	80 75 5	0
(1a-2)+(B-16) 40 + 36	60 50 10	50 50 0	50 44 6	90 88 2	90 88 2	90 90 0	0
(1a-2)+(B-17) 40 + 16	80 65 15	60 40 20	50 44 6	60 52 8	70 68 2	60 55 5	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 5 Follar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-3)+(8-1) 20 + 250	100 30 70	100 40 60	90 64 26	90 30 60	100 40 60	90 30 60	0
(1a-3)+(8-10) 40 + 250	100 100 0	100 76 24	70 51 19	60 50 10	80 60 20	60 40 20	0
(1a-3)+(8-7) 40 + 250	100 90 10	100 76 24	100 58 42	60 50 10	70 60 10	60 40 20	0
(1a-3)+(8-6) 40 + 250	100 95 5	100 60 40	100 30 70	80 50 30	80 60 20	90 40 50	0
(1a-3)+(8-20) 40 + 250	50 50 0	70 64 6	50 30 20	60 50 10	60 60 0	60 40 20	0
(1a-3)+(8-3) 40 + 62	100 95 5	100 96 4	90 72 18	90 50 40	100 76 24	100 40 60	0
(1a-3)+(8-2) 20 + 125	100 86 14	90 40 50	100 10 90	90 58 32	100 76 24	100 30 70	0
(1a-3)+(8-4) 40 + 125	100 60 40	100 60 40	90 30 60	90 60 30	100 84 16	90 40 50	0
(1a-3)+(8-9) 40 + 250	100 100 0	100 68 32	80 44 36	50 50 0	70 60 10	50 40 10	0
(1a-3)+(8-14) 40 + 16	70 60 10	70 68 2	50 44 6	80 80 0	80 80 0	80 76 4	0
(1a-3)+(8-15) 40 + 10	70 60 10	70 68 2	50 44 6	80 75 5	80 80 0	80 70 10	0
(1a-3)+(8-16) 40 + 36	60 50 10	60 60 0	50 44 6	90 90 0	90 88 2	90 88 2	0
(1a-3)+(8-17) 40 + 16	80 65 15	70 68 2	50 44 6	60 60 0	70 68 2	50 46 4	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)
A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 6 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-4)+(B-1) 20 + 250	100: 40: 60	100: 30: 70	90: 64: 26	90: 20: 70	100: 30: 70	100: 20: 80	0
(1a-4)+(B-10) 40 + 250	100: 100: 0	100: 70: 30	80: 51: 29	60: 40: 20	80: 50: 30	60: 40: 20	0
(1a-4)+(B-7) 40 + 250	100: 90: 10	100: 70: 30	100: 58: 42	60: 40: 20	70: 50: 20	70: 40: 30	0
(1a-4)+(B-6) 40 + 250	100: 95: 5	100: 50: 50	100: 30: 70	80: 40: 40	80: 50: 30	80: 40: 40	0
(1a-4)+(B-20) 40 + 250	70: 50: 20	60: 55: 5	50: 30: 20	60: 40: 20	60: 50: 10	50: 40: 10	0
(1a-4)+(B-3) 40 + 62	100: 95: 5	100: 95: 5	90: 72: 18	90: 40: 50	100: 70: 30	90: 40: 50	0
(1a-4)+(B-2) 20 + 125	100: 88: 12	90: 30: 60	100: 10: 90	90: 52: 38	100: 72: 28	100: 20: 80	0
(1a-4)+(B-4) 40 + 125	100: 60: 40	100: 50: 50	100: 30: 70	90: 52: 38	100: 80: 20	100: 40: 60	0
(1a-4)+(B-9) 40 + 250	100: 100: 0	100: 60: 40	90: 44: 48	50: 40: 10	60: 50: 10	50: 40: 10	0
(1a-4)+(B-14) 40 + 16	70: 60: 10	70: 60: 10	50: 44: 6	80: 76: 4	80: 75: 5	80: 76: 4	0
(1a-4)+(B-15) 40 + 10	70: 60: 10	60: 60: 0	50: 44: 6	70: 70: 0	80: 75: 5	80: 70: 10	0
(1a-4)+(B-16) 40 + 36	60: 50: 10	60: 50: 10	50: 44: 6	90: 88: 2	90: 85: 5	90: 88: 2	0
(1a-4)+(B-17) 40 + 16	80: 65: 15	70: 60: 10	60: 44: 16	60: 52: 8	70: 60: 10	50: 46: 4	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 7 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-5)+(B-1) 20 + 250	100: 40: 60	100: 30: 70	90: 64: 26	90: 20: 70	100: 30: 70	90: 20: 70	0
(1a-5)+(B-10) 40 + 250	100: 100: 0	100: 70: 30	90: 51: 39	60: 40: 20	70: 50: 20	60: 30: 30	0
(1a-5)+(B-7) 40 + 250	100: 92: 8	100: 70: 30	100: 58: 42	60: 40: 20	60: 50: 10	70: 30: 40	0
(1a-5)+(B-6) 40 + 250	100: 96: 4	100: 50: 50	100: 30: 70	70: 40: 30	80: 50: 30	80: 30: 50	0
(1a-5)+(B-20) 40 + 250	60: 60: 0	60: 55: 5	50: 30: 20	50: 40: 10	60: 50: 10	50: 30: 20	0
(1a-5)+(B-3) 40 + 62	100: 96: 4	100: 95: 5	90: 72: 18	100: 40: 60	100: 70: 30	100: 30: 70	0
(1a-5)+(B-2) 20 + 125	100: 68: 32	100: 50: 50	90: 30: 60	90: 52: 38	100: 80: 20	100: 30: 70	0
(1a-5)+(B-4) 40 + 125	100: 68: 32	100: 50: 50	90: 30: 60	90: 52: 38	100: 80: 20	100: 30: 70	0
(1a-5)+(B-9) 40 + 250	100: 100: 0	100: 60: 40	90: 44: 46	60: 40: 20	60: 50: 10	50: 30: 20	0
(1a-5)+(B-14) 40 + 16	70: 68: 2	70: 60: 10	50: 44: 6	80: 76: 4	80: 75: 5	80: 72: 8	0
(1a-5)+(B-15) 40 + 10	70: 68: 2	70: 60: 10	50: 44: 6	70: 70: 0	80: 75: 5	70: 65: 5	0
(1a-5)+(B-16) 40 + 36	70: 60: 10	70: 50: 20	50: 44: 6	90: 88: 2	90: 85: 5	90: 88: 4	0
(1a-5)+(B-17) 40 + 16	80: 72: 8	70: 60: 10	70: 44: 26	60: 52: 8	70: 60: 10	50: 37: 13	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 8 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-7)+(B-1) 20 + 250	100: 30: 70	100: 30: 70	100: 64: 36	90: 20: 70	100: 20: 80	90: 20: 70	0
(1a-7)+(B-10) 40 + 250	100: 100: 0	100: 70: 30	90: 58: 32	60: 50: 10	70: 50: 20	60: 40: 20	0
(1a-7)+(B-7) 40 + 250	100: 88: 12	100: 70: 30	100: 64: 36	60: 50: 10	70: 50: 20	70: 40: 30	0
(1a-7)+(B-6) 40 + 250	100: 94: 6	100: 50: 50	100: 40: 60	80: 50: 30	80: 50: 30	70: 40: 30	0
(1a-7)+(B-20) 40 + 250	60: 40: 20	60: 55: 5	50: 40: 10	60: 50: 10	60: 50: 10	50: 40: 10	0
(1a-7)+(B-3) 40 + 62	100: 94: 6	100: 95: 5	90: 76: 14	100: 50: 50	100: 70: 30	100: 40: 60	0
(1a-7)+(B-2) 20 + 125	100: 86: 14	100: 30: 70	100: 30: 70	100: 52: 48	100: 68: 32	90: 20: 70	0
(1a-7)+(B-4) 40 + 125	100: 52: 48	100: 50: 50	90: 40: 50	90: 60: 30	100: 80: 20	90: 40: 50	0
(1a-7)+(B-9) 40 + 250	100: 100: 0	100: 60: 40	90: 52: 38	60: 50: 10	60: 50: 10	50: 40: 10	0
(1a-7)+(B-14) 40 + 16	60: 52: 8	70: 60: 10	60: 52: 8	80: 80: 0	80: 75: 5	80: 75: 4	0
(1a-7)+(B-15) 40 + 10	70: 52: 18	70: 60: 10	60: 52: 8	80: 50: 30	80: 50: 30	70: 70: 0	0
(1a-7)+(B-16) 40 + 36	50: 40: 10	70: 50: 20	60: 52: 8	90: 90: 0	90: 85: 5	90: 88: 2	0
(1a-7)+(B-17) 40 + 16	80: 58: 22	80: 60: 20	70: 52: 18	60: 60: 0	70: 60: 10	50: 46: 4	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Tables 3 ~ 8 show that all the compositions containing Compound (1a-1) and one of Compounds (B-1), (B-6), (B-7), (B-10) and (B-20) in combination and all the compositions containing one of compounds (1a-2) ~ (1a-5) and (1a-7) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing compound (1a-1) and one of Compounds (B-1), (B-10), (B-6) and (B-20),

the composition containing Compound (1a-1) and Compound (B-1) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (1a-1) and Compound (B-10) showed a high synergistic effect on velvet leaf and barnyard grass in particular,

the composition containing Compound (1a-1) and Compound (B-7) showed a high synergistic effect on slender amaranth and green foxtail in particular,

the composition containing Compound (Ia-1) and Compound (B-6) showed a high synergistic effect on velvet leaf and slender amaranth in particular, and
the composition containing Compound (Ia-1) and Compound (B-20) showed a high synergistic effect on velvet leaf in particular.

Further, the compositions containing compound (Ia-1) and compound (B-1), (B-10) or (B-6) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

In the compositions containing Compound (Ia-2) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-2) and compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-2) and Compound (B-10) showed a high synergistic effect on green foxtail in particular,
the composition containing Compound (Ia-2) and Compound (B-7) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ia-2) and Compound (B-6) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-2) and Compound (B-20) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-2) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular
the composition containing Compound (Ia-2) and Compound (B-2) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-2) and Compound (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-2) and Compound (B-9) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ia-2) and Compound (B-14) showed a high synergistic effect on cocklebur in particular,
the composition containing compound (Ia-2) and Compound (B-15) showed a high synergistic effect on cocklebur and green foxtail in particular,
the composition containing Compound (Ia-2) and Compound (B-16) showed a high synergistic effect on cocklebur in particular, and
the composition containing Compound (Ia-2) and Compound (B-17) showed a high synergistic effect on cocklebur and velvet leaf in particular.

In the compositions containing Compound (Ia-3) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-3) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-10) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ia-3) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-3) and Compound (B-6) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-20) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-4) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-9) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-3) and Compound (B-14) showed a high synergistic effect on cocklebur

in particular,

the composition containing Compound (Ia-3) and Compound (B-15) showed a high synergistic effect on cocklebur and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-16) showed a high synergistic effect on cocklebur in particular, and

the composition containing Compound (Ia-3) and Compound (B-17) showed a high synergistic effect on cocklebur in particular.

In the compositions containing Compound (Ia-4) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-4) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-4) and Compound (B-10) showed a high synergistic effect on velvet leaf, slender amaranth and crabgrass in particular,

the composition containing Compound (Ia-4) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ia-4) and Compound (B-6) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-4) and Compound (B-20) showed a high synergistic effect on cocklebur, slender amaranth and green foxtail in particular,

the composition containing Compound (Ia-4) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,

the composition containing Compound (Ia-4) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-4) and Compound (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-4) and Compound (B-9) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-4) and Compound (B-14) showed a high synergistic effect on cocklebur in particular,

the composition containing Compound (Ia-4) and Compound (B-15) showed a high synergistic effect on cocklebur and barnyard grass in particular,

the composition containing Compound (Ia-4) and Compound (B-16) showed a high synergistic effect on cocklebur and velvet leaf in particular, and

the composition containing Compound (Ia-4) and Compound (B-17) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing Compound (Ia-5) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-5) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-10) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-7) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-20) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-9) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-5) and Compound (B-14) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-5) and Compound (B-15) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-5) and Compound (B-16) showed a high synergistic effect on velvet leaf in particular, and

the composition containing Compound (Ia-5) and Compound (B-17) showed a high synergistic effect on slender amaranth in particular.

In the compositions containing Compound (Ia-7) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-7) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-10) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-7) and Compound (B-7) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth in particular,

the composition containing Compound (Ia-7) and Compound (B-20) showed a high synergistic effect on cocklebur in particular,

the composition containing Compound (Ia-7) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-9) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-7) and Compound (B-14) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-7) and Compound (B-15) showed a high synergistic effect on crabgrass in particular,

the composition containing Compound (Ia-7) and Compound (B-16) showed a high synergistic effect on velvet leaf in particular, and

the composition containing Compound (Ia-7) and Compound (B-17) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular.

Table 9 shows the results of foliar treatment tests of single active ingredients of Compounds (Ib-1), (Ib-2), (Ib-3) and Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17).

Tables 10 to 12 show the results of foliar treatment tests of compositions containing one of Compounds (Ib-1), (Ib-2) and (Ib-3) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17).

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Table 9

Foliar treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(lb-1)	40	60	20	20	40	60	40	0
	20	40	10	20	20	40	20	0
(lb-2)	40	50	30	30	40	60	40	0
	20	30	20	20	20	40	20	0
(lb-3)	40	50	30	20	50	60	40	0
	20	40	20	20	20	40	30	0
(B-1)	250	20	0	0	0	0	0	0
(B-2)	125	80	0	0	40	60	0	0
(B-3)	62	90	90	60	0	40	0	0
(B-4)	125	20	0	0	20	60	0	0
(B-5)	62	20	20	0	0	0	0	0
(B-6)	250	20	0	20	0	0	0	0
(B-7)	125	90	90	40	0	0	0	0
(B-8)	250	90	0	0	0	0	0	0
(B-9)	250	90	20	40	0	0	0	0
(B-10)	250	90	80	0	0	20	0	0
(B-14)	16	20	20	20	60	50	60	0
(B-15)	10	20	20	20	50	50	50	0
(B-16)	36	0	0	20	80	70	80	0
(B-17)	16	30	20	20	20	20	10	0
(a): Cocklebur (b): Velvet leaf (c): Slender amaranth (d): Green foxtail (e): Crabgrass (f): Barnyard grass								

Table 10 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1b-1)+(B-1) 20 + 250	80 52 28	100 10 90	60 20 40	100 20 80	100 40 60	80 20 60	0
(1b-1)+(B-2) 20 + 125	100 88 12	100 10 90	80 20 60	100 52 48	100 76 24	80 20 60	0
(1b-1)+(B-3) 40 + 62	100 96 4	100 92 8	100 68 32	100 40 60	100 76 24	100 40 60	0
(1b-1)+(B-4) 40 + 125	100 68 32	100 20 80	80 20 60	90 52 38	100 84 16	100 40 60	0
(1b-1)+(B-5) 20 + 62	100 52 48	100 28 72	40 20 20	80 20 60	80 40 40	80 20 60	0
(1b-1)+(B-6) 40 + 250	100 68 32	100 20 80	100 36 64	90 40 50	100 60 40	90 40 50	0
(1b-1)+(B-7) 40 + 125	100 96 4	100 92 8	100 52 48	40 40 0	60 60 0	40 40 0	0
(1b-1)+(B-8) 20 + 250	100 90 10	100 20 80	60 20 40	100 40 60	100 60 40	80 40 40	0
(1b-1)+(B-9) 40 + 250	100 96 4	80 36 44	100 36 64	60 40 20	80 60 20	40 40 0	0
(1b-1)+(B-10) 40 + 250	100 96 4	100 84 16	40 20 20	90 40 50	90 68 22	80 40 40	0
(1b-1)+(B-14) 40 + 16	80 68 12	60 36 24	40 36 4	80 76 4	80 80 0	80 76 4	0
(1b-1)+(B-15) 40 + 10	70 68 2	40 36 4	40 36 4	80 70 10	80 80 0	80 70 10	0
(1b-1)+(B-16) 40 + 36	70 60 10	40 20 20	40 36 4	90 88 2	90 88 2	90 88 2	0
(1b-1)+(B-17) 40 + 16	80 72 8	40 36 4	40 36 4	60 52 8	70 68 2	50 48 2	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 11 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1b-2)+(B-1) 20 + 250	90: 44: 46	100: 20: 80	60: 20: 40	90: 20: 70	100: 40: 60	90: 20: 70	0
(1b-2)+(B-2) 20 + 125	100: 86: 14	90: 20: 70	90: 20: 70	90: 52: 38	100: 76: 24	80: 20: 60	0
(1b-2)+(B-3) 40 + 62	100: 95: 5	100: 93: 7	100: 72: 28	100: 40: 60	100: 76: 24	100: 40: 60	0
(1b-2)+(B-4) 40 + 125	100: 60: 40	100: 30: 70	80: 30: 50	100: 52: 48	90: 84: 6	100: 40: 60	0
(1b-2)+(B-5) 20 + 62	100: 44: 56	90: 36: 54	40: 20: 20	80: 20: 60	90: 40: 50	80: 20: 60	0
(1b-2)+(B-6) 40 + 250	100: 60: 40	90: 30: 60	100: 44: 56	90: 40: 50	100: 60: 40	90: 40: 50	0
(1b-2)+(B-7) 40 + 125	100: 95: 5	100: 93: 7	90: 56: 32	60: 40: 20	80: 60: 20	60: 40: 20	0
(1b-2)+(B-8) 20 + 250	100: 95: 5	100: 30: 70	60: 20: 40	90: 40: 50	90: 60: 30	90: 40: 50	0
(1b-2)+(B-9) 40 + 250	100: 95: 5	90: 44: 46	100: 44: 56	60: 40: 20	80: 60: 20	40: 40: 0	0
(1b-2)+(B-10) 40 + 250	100: 95: 5	100: 86: 14	50: 30: 20	90: 40: 50	90: 58: 22	90: 40: 50	0
(1b-2)+(B-14) 40 + 16	60: 60: 0	50: 44: 6	50: 44: 6	80: 75: 4	80: 80: 0	80: 76: 4	0
(1b-2)+(B-15) 40 + 10	70: 60: 10	50: 44: 6	70: 44: 26	80: 70: 10	80: 80: 0	80: 70: 10	0
(1b-2)+(B-16) 40 + 36	60: 50: 10	40: 30: 10	50: 44: 6	90: 88: 2	90: 88: 2	90: 88: 2	0
(1b-2)+(B-17) 40 + 16	80: 65: 15	50: 44: 6	50: 44: 6	50: 52: 8	70: 68: 2	50: 46: 4	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 12 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(lb-3)+(B-1) 20 + 250	90, 52, 38	100, 20, 80	70, 20, 50	90, 20, 70	100, 40, 60	100, 30, 70	0
(lb-3)+(B-2) 20 + 125	100, 88, 12	100, 20, 80	100, 20, 80	100, 52, 48	100, 76, 24	80, 30, 50	0
(lb-3)+(B-3) 40 + 62	100, 95, 5	100, 93, 7	100, 68, 32	100, 50, 50	100, 76, 24	90, 40, 50	0
(lb-3)+(B-4) 40 + 125	100, 60, 40	90, 30, 60	90, 20, 70	90, 60, 30	100, 84, 16	100, 40, 60	0
(lb-3)+(B-5) 20 + 62	90, 52, 38	90, 36, 54	40, 20, 20	90, 20, 70	90, 40, 50	90, 30, 60	0
(lb-3)+(B-6) 40 + 250	100, 60, 40	90, 30, 60	100, 36, 64	100, 50, 50	100, 60, 40	90, 40, 50	0
(lb-3)+(B-7) 40 + 125	100, 95, 5	100, 93, 7	70, 52, 18	70, 50, 20	80, 60, 20	70, 40, 30	0
(lb-3)+(B-8) 20 + 250	100, 95, 5	100, 30, 70	60, 20, 40	100, 50, 50	100, 60, 40	80, 40, 40	0
(lb-3)+(B-9) 40 + 250	100, 95, 5	90, 44, 46	100, 36, 64	60, 50, 10	80, 60, 20	70, 40, 30	0
(lb-3)+(B-10) 40 + 250	100, 95, 5	100, 86, 14	80, 20, 60	90, 50, 40	90, 60, 30	80, 40, 40	0
(lb-3)+(B-14) 40 + 16	60, 60, 0	50, 44, 6	40, 36, 4	80, 80, 0	80, 86, 0	80, 76, 4	0
(lb-3)+(B-15) 40 + 10	70, 60, 10	50, 44, 6	40, 36, 4	80, 75, 5	90, 80, 10	80, 70, 10	0
(lb-3)+(B-16) 40 + 36	70, 50, 20	50, 30, 20	50, 36, 14	90, 90, 0	90, 88, 2	90, 88, 2	0
(lb-3)+(B-17) 40 + 16	80, 65, 15	50, 44, 6	40, 36, 4	70, 60, 10	70, 68, 2	50, 46, 4	0

(a): Cocklebur (d): Green foxtail
 (b): Velvet leaf (e): Crabgrass
 (c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)
 A larger difference (Δ) means a larger synergistic effect
 due to the combined use of active ingredients.

Tables 10, 11 and 12 show that all the compositions containing Compound one of Compounds (lb-1), (lb-2) and (lb-3) and one of compounds (B-1) ~ (B-10) and (B-14) ~ (B-17) in combination showed high synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing Compound (lb-1) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Compound (lb-1) and Compound (B-1) showed a high synergistic effect on velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (lb-1) and Compound (B-2) or (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (lb-1) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,

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the composition containing Compound (Ib-1) and Compound (B-5) showed a high synergistic effect on velvet leaf, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-1) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
5 the composition containing Compound (Ib-1) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-1) and Compound (B-8) showed a high synergistic effect on velvet leaf and green foxtail in particular,
the composition containing Compound (Ib-1) and Compound (B-9) showed a high synergistic effect on slender
10 amaranth in particular,
the composition containing Compound (Ib-1) and Compound (B-10) showed a high synergistic effect on green foxtail in particular, and
the composition containing Compound (Ib-1) and Compound (B-14) or (B-16) showed a high synergistic effect on velvet leaf in particular.

15 In the compositions containing Compound (Ib-2) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Composition (Ib-2) and Compound (B-1) showed a high synergistic effect on velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
20 the composition containing Compound (Ib-2) and Compound (B-2) or (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-5) showed a high synergistic effect on cocklebur,
25 velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,
30 the composition containing Compound (Ib-2) and Compound (B-8) showed a high synergistic effect on velvet leaf, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-9) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-2) and Compound (B-10) showed a high synergistic effect on green foxtail and barnyard grass in particular, and
35 the composition containing Compound (Ib-2) and Compound (B-15) showed a high synergistic effect on slender amaranth in particular.

40 In the compositions containing Compound (Ib-3) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ib-3) and compound (B-1) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-2) or (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
45 the composition containing Compound (Ib-3) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-5) showed a high synergistic effect on velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
50 the composition containing Compound (Ib-3) and Compound (B-7) showed a high synergistic effect on barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-8) showed a high synergistic effect on velvet leaf and green foxtail in particular,
55 the composition containing Compound (Ib-3) and Compound (B-9) or (B-10) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-3) and Compound (B-16) showed a high synergistic effect on cocklebur and velvet leaf in particular, and
the composition containing Compound (Ib-3) and Compound (B-17) showed a high synergistic effect on cocklebur

in particular.

Further, the compositions containing Compound (Ib-1), (Ib-2) or (Ib-3) and one of Compounds (B-1) ~ (B-6) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

Table 13 shows the results of foliar treatment tests on single active ingredients of Compound (Ic-1) and Compounds (B-1) ~ (B-4), (B-6) ~ (B-10) and (B-14) ~ (B-17).

Table 14 shows the results of foliar treatment tests on compositions of Compound (Ic-1) and one of Compounds (B-1) ~ (B-4), (B-6) ~ (B-10) and (B-14) ~ (B-17).

Table 13

Foliar treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ic-1)	40	20	20	0	20	20	20	0
	20	0	0	0	0	0	0	0
(B-1)	250	0	20	40	0	20	0	0
(B-2)	250	0	0	0	20	80	0	0
(B-3)	125	80	80	80	80	20	80	0
(B-4)	500	80	90	60	40	60	0	0
(B-6)	250	0	0	0	0	0	0	0
(B-7)	125	80	90	40	0	0	0	0
(B-8)	125	90	0	0	0	0	0	0
(B-9)	125	90	0	40	0	0	0	0
(B-10)	125	20	10	0	0	20	0	0
(B-14)	16	20	20	20	60	50	60	0
(B-15)	10	10	10	20	50	60	50	0
(B-16)	36	0	0	20	80	70	80	0
(B-17)	16	30	20	20	20	20	10	0
(a): Cocklebur (b): Velvet leaf (c): Slender amaranth (d): Green foxtail (e): Crabgrass (f): Barnyard grass								

Table 14 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
(1c-1)+(B-1)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	corn
40 + 250	100, 20, 80	100, 20, 80	60, 40, 20	40, 20, 20	100, 36, 64	30, 20, 10	
(1c-1)+(B-2)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 250	100, 20, 80	100, 20, 80	20, 0, 20	80, 36, 44	100, 80, 20	80, 20, 80	
(1c-1)+(B-3)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 125	100, 84, 16	100, 84, 16	100, 80, 20	80, 20, 60	40, 20, 20	80, 20, 60	
(1c-1)+(B-4)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 500	100, 84, 16	100, 92, 8	100, 60, 40	90, 52, 38	100, 68, 32	60, 20, 40	
(1c-1)+(B-6)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 250	100, 20, 80	100, 20, 80	20, 0, 20	20, 20, 0	100, 20, 80	20, 20, 0	
(1c-1)+(B-7)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 125	100, 84, 16	100, 92, 8	100, 40, 60	50, 20, 30	20, 20, 0	20, 20, 0	
(1c-1)+(B-8)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 125	100, 92, 8	20, 20, 0	100, 0, 100	60, 20, 40	60, 20, 40	40, 20, 20	
(1c-1)+(B-9)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
20 + 125	100, 90, 10	40, 0, 40	100, 40, 60	0, 0, 0	20, 0, 20	0, 0, 0	
(1c-1)+(B-10)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 125	100, 36, 64	100, 28, 72	20, 0, 20	60, 20, 40	80, 36, 44	60, 20, 40	
(1c-1)+(B-14)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 16	40, 36, 4	40, 36, 4	40, 20, 20	70, 68, 2	80, 60, 20	70, 68, 2	
(1c-1)+(B-15)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 10	60, 27, 33	50, 27, 23	40, 20, 20	70, 60, 10	80, 62, 18	80, 60, 20	
(1c-1)+(B-16)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 36	60, 20, 40	40, 20, 20	50, 20, 30	90, 84, 6	90, 76, 14	90, 84, 6	
(1c-1)+(B-17)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	(F):(E);(Δ)	
40 + 16	80, 44, 36	40, 36, 4	40, 20, 20	60, 36, 24	70, 36, 34	50, 28, 22	

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

All the compositions containing Compound (1c-1) and one of Compounds (B-1) ~ (B-4), (B-6) ~ (B-10) and (B-14) ~ (B-17) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing Compound (1c-1) and one of Compounds B-1) ~ (B-4), (B-6) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Compound (1c-1) and Compound (B-1) or (B-6) showed a high synergistic effect on cocklebur, velvet leaf and crabgrass in particular,

the composition containing Compound (1c-1) and Compound (B-) showed a high synergistic effect on in particular, the composition containing Compound (1c-1) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf and barnyard grass in particular,

the composition containing Compound (1c-1) and Compound (B-3) showed a high synergistic effect on green foxtail

and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-4) showed a high synergistic effect on slender amaranth, green foxtail and barnyard grass in particular,

the composition containing Compound (Ic-1) and one of Compounds (B-7) ~ (B-9) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ic-1) and Compound (B-10) showed a high synergistic effect on cocklebur and velvet leaf in particular,

the composition containing Compound (Ic-1) and Compound (B-14) showed a high synergistic effect on slender amaranth and crabgrass in particular,

the composition containing Compound (Ic-1) and Compound (B-15) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-16) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and

the composition containing Compound (Ic-1) and Compound (B-17) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail, crabgrass and barnyard grass in particular.

Further, the compositions containing Compound (Ic-1) and one of Compounds (B-1) ~ (B-4), (B-6) and (B-10) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

Example 2 [Soil treatment test]

Seeds of weeds such as cocklebur, velvet leaf, Slender amaranth, green foxtail, crabgrass and barnyard grass and seeds of corn were sown in 1/2,000-are Wagner pots filled with upland soil, and covered with upland soil. One day after the seeds were sown, a predetermined amount of the herbicide obtained in the same manner as in Preparation Example 5 except that one of Compounds (Ia-2) ~ (Ia-5), (Ia-7), (Ib-1) ~ (Ib-3), (Ic-1) and (Ic-2) was suspended in water and uniformly sprayed in a solution amount of 1,000 liters/hectare.

Twenty days after the treatment, the herbicide was determined for phytotoxicity to crops and herbicidal efficacy in the same manner as in the foliar treatment test. The herbicidal efficacy (weed control ratio %) was determined on the basis of the above equation (A).

Table 15 shows the results of soil treatment tests on single active ingredients of Compounds (Ia-2) ~ (Ia-5) and Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 16 to 20 show the results of soil treatment tests on compositions of one of Compounds (Ia-2) ~ (Ia-5) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

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Table 15

Soil treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ia-2)	80	20	80	20	20	20	40	0
	40	0	60	0	10	20	40	0
(Ia-3)	80	40	20	0	40	20	40	0
	40	20	20	0	10	20	30	0
(Ia-4)	80	40	60	0	40	50	30	0
	40	20	30	0	20	20	20	0
(Ia-5)	80	40	60	0	30	50	40	0
	40	20	20	0	20	30	20	0
(Ia-7)	80	40	50	40	40	50	40	0
	40	20	30	10	20	30	30	0
(B-7)	125	90	40	0	0	0	0	0
(B-3)	125	90	90	90	20	0	20	0
(B-2)	125	20	0	0	0	0	0	0
(B-4)	500	0	20	20	0	0	0	0
(B-12)	500	20	0	0	60	60	80	0
(B-13)	250	0	20	20	80	80	80	0
(B-11)	250	0	0	0	60	80	40	0
(B-18)	250	20	10	40	70	70	70	0
(B-19)	250	0	0	10	50	70	60	0
(a): Cocklebur (b): Velvet leaf (c): Slender amaranth (d): Green foxtail (e): Crabgrass (f): Barnyard grass								

Table 16 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-2)+(B-7) 40 + 125	100: 90: 10	100: 76: 24	100: 0: 100	20: 10: 10	100: 20: 80	80: 20: 60	0
(1a-2)+(B-3) 40 + 125	100: 90: 10	100: 95: 5	100: 90: 10	100: 36: 64	100: 20: 80	100: 52: 48	0
(1a-2)+(B-2) 80 + 125	100: 36: 64	100: 80: 20	100: 20: 80	40: 20: 20	100: 20: 80	100: 40: 60	0
(1a-2)+(B-4) 40 + 500	90: 20: 70	100: 84: 16	100: 20: 80	40: 20: 20	60: 20: 40	100: 40: 60	0
(1a-2)+(B-12) 80 + 500	80: 36: 44	100: 20: 80	100: 20: 80	90: 68: 22	90: 68: 22	100: 98: 2	0
(1a-2)+(B-13) 80 + 250	20: 20: 0	100: 84: 16	100: 36: 64	20: 10: 10	100: 20: 80	80: 40: 40	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 17 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-3)+(B-7) 40 + 125	100: 92: 8	100: 52: 48	40: 0: 40	20: 10: 10	60: 20: 40	60: 30: 30	0
(1a-3)+(B-3) 40 + 125	100: 92: 8	100: 92: 8	100: 90: 10	90: 28: 62	100: 20: 80	100: 44: 56	0
(1a-3)+(B-2) 80 + 125	100: 52: 48	100: 20: 80	80: 0: 80	80: 40: 40	90: 20: 70	90: 40: 50	0
(1a-3)+(B-4) 40 + 500	80: 20: 60	90: 36: 54	100: 20: 80	80: 10: 70	90: 20: 70	100: 30: 70	0
(1a-3)+(B-12) 80 + 500	80: 52: 28	80: 20: 60	60: 0: 60	90: 76: 14	100: 68: 32	100: 94: 6	0
(1a-3)+(B-13) 80 + 250	50: 40: 10	80: 36: 44	80: 20: 60	100: 88: 12	100: 84: 16	100: 94: 6	0
(1a-3)+(B-11) 80 + 250	60: 40: 20	90: 20: 70	80: 0: 80	100: 76: 24	100: 84: 16	100: 64: 36	0
(1a-3)+(B-18) 80 + 250	80: 52: 28	80: 28: 52	80: 40: 40	100: 82: 18	100: 76: 24	100: 82: 18	0
(1a-3)+(B-19) 80 + 250	80: 40: 40	80: 20: 60	90: 10: 80	100: 70: 30	100: 76: 24	100: 76: 24	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 18 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
(1a-4)+(B-7) 40 + 125	100	92	8	100	58	42	0
(1a-4)+(B-3) 40 + 125	100	92	8	100	93	7	0
(1a-4)+(B-2) 80 + 125	100	52	48	100	60	40	0
(1a-4)+(B-4) 40 + 500	90	20	70	90	44	46	0
(1a-4)+(B-12) 80 + 500	90	52	38	90	60	30	0
(1a-4)+(B-13) 80 + 250	50	40	10	80	68	12	0
(1a-4)+(B-11) 80 + 250	70	30	40	100	60	40	0
(1a-4)+(B-18) 80 + 250	90	52	38	100	64	36	0
(1a-4)+(B-19) 80 + 250	90	40	50	80	60	20	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 19 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-5)+(B-7) 40 + 125	100; 92; 8	100; 52; 48	70; 0; 70	40; 20; 20	50; 30; 20	40; 20; 20	0
(1a-5)+(B-3) 40 + 125	100; 92; 8	100; 92; 8	90; 90; 10	80; 36; 44	100; 30; 70	100; 36; 64	0
(1a-5)+(B-2) 80 + 125	100; 52; 48	100; 60; 40	80; 60; 20	90; 30; 60	100; 50; 50	90; 40; 50	0
(1a-5)+(B-4) 40 + 500	100; 20; 80	100; 36; 64	100; 20; 80	90; 20; 70	90; 30; 60	100; 20; 80	0
(1a-5)+(B-12) 80 + 500	100; 52; 48	100; 60; 40	60; 0; 60	90; 72; 18	100; 80; 20	100; 94; 6	0
(1a-5)+(B-13) 80 + 250	50; 40; 10	70; 68; 2	40; 20; 20	100; 86; 14	100; 90; 10	100; 94; 6	0
(1a-5)+(B-11) 80 + 250	90; 40; 50	100; 60; 40	40; 0; 40	100; 72; 28	100; 90; 10	100; 64; 36	0
(1a-5)+(B-18) 80 + 250	90; 52; 38	90; 64; 26	80; 40; 40	100; 79; 21	100; 85; 15	100; 82; 18	0
(1a-5)+(B-19) 80 + 250	80; 40; 40	90; 60; 30	100; 10; 90	100; 65; 35	100; 85; 15	100; 76; 24	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 20 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1a-7)+(B-7) 40 + 125	100: 92: 8	100: 58: 42	80: 10: 70	50: 20: 30	50: 30: 20	90: 20: 70	0
(1a-7)+(B-3) 40 + 125	100: 92: 8	100: 93: 7	100: 91: 9	80: 36: 44	100: 30: 70	100: 44: 56	0
(1a-7)+(B-2) 80 + 125	100: 52: 48	100: 50: 50	80: 20: 60	90: 40: 50	100: 50: 50	100: 40: 60	0
(1a-7)+(B-4) 40 + 500	90: 20: 70	100: 44: 56	100: 36: 64	90: 20: 70	100: 30: 70	100: 30: 70	0
(1a-7)+(B-12) 80 + 500	100: 52: 48	100: 50: 50	80: 20: 60	100: 76: 24	100: 80: 20	100: 94: 6	0
(1a-7)+(B-13) 80 + 250	60: 40: 20	80: 60: 20	80: 36: 44	100: 88: 12	100: 90: 10	100: 94: 6	0
(1a-7)+(B-11) 80 + 250	70: 40: 30	90: 50: 40	60: 20: 40	100: 76: 24	100: 90: 10	100: 64: 36	0
(1a-7)+(B-18) 80 + 250	80: 52: 28	90: 55: 35	80: 52: 28	100: 82: 18	100: 85: 15	100: 82: 18	0
(1a-7)+(B-19) 80 + 250	100: 40: 60	90: 50: 40	100: 28: 72	100: 70: 30	100: 85: 15	100: 76: 24	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 16 ~ 20 show that all the compositions containing Compound (1a-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-12) and (B-13) in combination and all the compositions containing one of Compounds (1a-2) ~ (1a-5) and (1a-7) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing (Ia-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-12) and (B-13),

the composition containing Compound (Ia-2) and Compound (B-7) showed a high synergistic effect on crabgrass, barnyard grass and slender amaranth in particular,
 5 the composition containing Compound (Ia-2) and Compound (B-) showed a high synergistic effect on in particular,
 the composition containing Compound (Ia-2) and Compound (B-3) showed a high synergistic effect on green fox-tail, crabgrass and barnyard grass in particular,
 the composition containing Compound (Ia-2) and Compound (B-2) showed a high synergistic effect on cocklebur, slender amaranth, crabgrass and barnyard grass in particular,
 10 the composition containing Compound (Ia-2) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth and barnyard grass in particular,
 the composition containing Compound (Ia-2) and Compound (B-12) showed a high synergistic effect on velvet leaf and slender amaranth in particular, and
 the composition containing Compound (Ia-2) and Compound (B-13) showed a high synergistic effect on slender amaranth and crabgrass in particular.

Further, the compositions containing Compound (Ia-2) and one of Compounds (B-2) ~ (B-4) and (B-7) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

In the compositions containing Compound (Ia-3) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-3) and Compound (B-7) showed a high synergistic effect on velvet leaf, slender amaranth and crabgrass in particular,
 the composition containing Compound (Ia-3) and Compound (B-3) showed a high synergistic effect on green fox-tail, crabgrass and barnyard grass in particular,
 25 the composition containing Compound (Ia-3) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, crabgrass and barnyard grass in particular,
 the composition containing Compound (Ia-3) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
 30 the composition containing Compound (Ia-3) and Compound (B-12) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
 the composition containing Compound (Ia-3) and Compound (B-13) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
 the composition containing Compound (Ia-3) and Compound (B-11) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
 35 the composition containing Compound (Ia-3) and Compound (B-18) showed a high synergistic effect on velvet leaf and slender amaranth in particular, and
 the composition containing Compound (Ia-3) and Compound (B-19) showed a high synergistic effect on velvet leaf and slender amaranth in particular.

In the compositions containing Compound (Ia-4) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-4) and Compound (B-7) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
 45 the composition containing Compound (Ia-4) and Compound (B-3) showed a high synergistic effect on green fox-tail, crabgrass and barnyard grass in particular,
 the composition containing Compound (Ia-4) and Compound (B-2) showed a high synergistic effect on cocklebur, slender amaranth, crabgrass and barnyard grass in particular,
 50 the composition containing Compound (Ia-4) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
 the composition containing Compound (Ia-4) and Compound (B-12) showed a high synergistic effect on slender amaranth in particular,
 the composition containing Compound (Ia-4) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular,
 55 the composition containing Compound (Ia-4) and Compound (B-11) showed a high synergistic effect on cocklebur, slender amaranth and barnyard grass in particular,
 the composition containing Compound (Ia-4) and Compound (B-18) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and

the composition containing Compound (Ia-4) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing Compound (Ia-5) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-5) and Compound (B-7) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-5) and Compound (B-3) showed a high synergistic effect on crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-2) showed a high synergistic effect on cocklebur, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-12) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-5) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ia-5) and Compound (B-11) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-5) and Compound (B-18) showed a high synergistic effect on cocklebur and slender amaranth in particular, and

the composition containing Compound (Ia-5) and Compound (B-19) showed a high synergistic effect on slender amaranth in particular.

In the compositions containing Compound (Ia-7) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-7) and Compound (B-7) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-3) showed a high synergistic effect on crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-12) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-7) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ia-7) and Compound (B-11) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-7) and Compound (B-18) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and

the composition containing Compound (Ia-7) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

Table 21 shows the results of soil treatment tests on single active ingredients of Compounds (Ib-1) ~ (Ib-3) and Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 22 to 24 show the results of soil treatment tests on compositions of one of Compounds (Ib-1) ~ (Ib-3) and one of compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

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Table 21

Soil treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ib-1)	80	20	60	20	40	80	40	0
	40	0	20	0	20	50	20	0
(Ib-2)	80	10	60	20	30	70	40	0
	40	0	20	0	0	20	0	0
(Ib-3)	80	20	50	20	30	60	40	0
(B-2)	125	0	0	0	0	0	0	0
(B-3)	62	90	90	60	0	40	0	0
(B-4)	250	0	20	20	0	0	0	0
(B-7)	62	40	20	0	0	0	0	0
(B-11)	250	0	0	0	60	80	40	0
(B-12)	125	0	0	0	20	60	60	0
(B-13)	125	0	0	0	80	80	20	0
(B-18)	250	20	10	40	70	70	70	0
(B-19)	250	0	0	10	50	70	60	0
(a): Cocklebur (b): Velvet leaf (c): Slender amaranth (d): Green foxtail (e): Crabgrass (f): Barnyard grass								

Table 22 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1b-1)+(B-2) 80 + 125	80: 20: 60	100: 60: 40	100: 20: 80	100: 40: 60	100: 80: 20	100: 40: 60	0
(1b-1)+(B-3) 40 + 62	100: 90: 10	100: 92: 8	100: 60: 40	40: 20: 20	100: 70: 30	90: 20: 70	0
(1b-1)+(B-4) 80 + 250	80: 20: 60	100: 68: 32	80: 36: 44	80: 40: 40	80: 40: 40	90: 80: 10	0
(1b-1)+(B-7) 80 + 62	100: 52: 48	100: 68: 32	20: 20: 0	80: 40: 40	90: 80: 10	80: 40: 40	0
(1b-1)+(B-11) 80 + 250	60: 20: 40	100: 60: 40	20: 20: 0	100: 76: 24	100: 96: 4	100: 64: 36	0
(1b-1)+(B-12) 80 + 250	60: 20: 40	100: 60: 40	100: 20: 80	100: 52: 48	100: 92: 8	100: 76: 24	0
(1b-1)+(B-13) 80 + 125	40: 20: 20	100: 60: 40	100: 20: 80	100: 88: 12	100: 96: 4	100: 52: 48	0
(1b-1)+(B-18) 80 + 250	80: 36: 44	80: 64: 16	90: 52: 38	90: 82: 8	100: 94: 6	100: 82: 18	0
(1b-1)+(B-19) 80 + 250	90: 20: 70	90: 60: 30	100: 28: 72	100: 70: 30	100: 94: 6	100: 76: 24	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 23 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1b-2)+(B-2) 80 + 125	80 10 70	100 60 40	90 20 70	100 30 70	100 70 30	100 40 60	0
(1b-2)+(B-3) 40 + 62	100 90 10	100 92 8	100 60 40	50 0 50	90 52 38	90 0 90	0
(1b-2)+(B-4) 80 + 250	80 10 70	100 68 32	90 36 54	90 30 60	90 70 20	90 40 50	0
(1b-2)+(B-7) 80 + 62	90 46 44	100 68 32	40 20 20	60 30 30	70 70 0	60 40 20	0
(1b-2)+(B-11) 80 + 250	50 10 40	100 60 40	40 20 20	100 72 28	100 94 6	100 64 36	0
(1b-2)+(B-12) 80 + 250	40 10 30	100 60 40	90 20 70	100 44 56	100 88 12	100 76 24	0
(1b-2)+(B-13) 80 + 125	50 10 40	100 60 40	100 20 80	100 86 14	100 94 6	100 52 48	0
(1b-2)+(B-18) 80 + 250	80 28 52	90 64 26	100 52 48	100 79 21	100 91 9	100 82 18	0
(1b-2)+(B-19) 80 + 250	90 10 80	90 60 30	100 28 72	100 65 35	100 91 9	100 76 24	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 24 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(1b-3)+(B-2) 80 + 125	90 20 70	100 50 50	90 20 70	100 30 70	100 60 40	100 40 60	0
(1b-3)+(B-3) 40 + 62	100 92 8	100 95 5	90 68 22	80 30 50	100 76 24	90 40 50	0
(1b-3)+(B-4) 80 + 250	80 20 60	100 60 40	90 36 54	80 30 50	80 60 20	90 40 50	0
(1b-3)+(B-7) 80 + 62	90 52 38	100 60 40	50 20 30	70 30 40	90 60 30	80 40 40	0
(1b-3)+(B-11) 80 + 250	60 20 40	100 50 50	60 20 40	100 72 28	100 92 8	100 64 36	0
(1b-3)+(B-12) 80 + 250	80 20 60	100 50 50	100 20 80	100 44 56	100 84 16	90 76 14	0
(1b-3)+(B-13) 80 + 125	70 20 50	100 50 50	100 20 80	100 86 14	100 92 8	100 52 48	0
(1b-3)+(B-18) 80 + 250	80 36 44	100 55 45	100 52 48	90 79 11	100 88 12	100 82 18	0
(1b-3)+(B-19) 80 + 250	80 20 60	90 50 40	100 28 72	100 65 35	100 88 12	100 76 24	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 22, 23 and 24 show that all the compositions containing one of Compounds (1b-1), (1b-2) and (1b-3) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing (1b-1) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18)

and (B-19),

the composition containing Compound (Ib-1) and Compound (B-2) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-1) and Compound (B-3) showed a high synergistic effect on barnyard grass in particular,
 the composition containing Compound (Ib-1) and Compound (B-4) showed a high synergistic effect on cocklebur in particular,
 the composition containing Compound (Ib-1) and Compound (B-7) showed a high synergistic effect on cocklebur in particular,
 the composition containing Compound (Ib-1) and Compound (B-11) showed a high synergistic effect on cocklebur, velvet leaf and barnyard grass in particular,
 the composition containing Compound (Ib-1) and Compound (B-12) or (B-13) showed a high synergistic effect on slender amaranth in particular, and
 the composition containing Compound (Ib-1) and Compound (B-18) or (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing (Ib-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ib-2) and Compound (B-2) or (B-4) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-2) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-2) and Compound (B-7) showed a high synergistic effect on cocklebur in particular,
 the composition containing Compound (Ib-2) and Compound (B-11) showed a high synergistic effect on cocklebur and velvet leaf in particular,
 the composition containing Compound (Ib-2) and Compound (B-12) showed a high synergistic effect on slender amaranth and green foxtail in particular,
 the composition containing Compound (Ib-2) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular, and
 the composition containing Compound (Ib-2) and Compound (B-18) or (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing (Ib-3) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ib-3) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-3) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-3) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-3) and Compound (B-7) showed a high synergistic effect on velvet leaf, green foxtail and barnyard grass in particular,
 the composition containing Compound (Ib-3) and Compound (B-11) showed a high synergistic effect on velvet leaf in particular,
 the composition containing Compound (Ib-3) and Compound (B-12) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and green foxtail in particular,
 the composition containing Compound (Ib-3) and Compound (B-13) or (B-18) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and
 the composition containing Compound (Ib-3) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

Table 25 shows the results of soil treatment tests on single active ingredients of Compounds (Ic-1) and (Ic-2) and Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 26 ~ 27 show the results of soil treatment tests on compositions of one of Compounds (Ic-1) and (Ic-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 25

Soil treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(lc-1)	80	50	60	0	30	40	30	0
(lc-2)	80	40	60	0	40	40	30	0
	40	0	40	0	0	20	0	0
(B-2)	250	60	0	0	0	20	0	0
(B-3)	125	90	90	90	0	40	40	0
(B-4)	500	0	0	0	0	0	0	0
(B-7)	125	80	20	20	0	0	0	0
(B-11)	250	0	0	0	60	80	80	0
(B-12)	250	0	0	0	60	60	90	0
(B-13)	250	0	0	0	90	90	90	0
(B-18)	250	20	10	40	70	70	70	0
(B-19)	250	0	0	10	50	70	60	0
(a): Cocklebur (b): Velvet leaf (c): Slender amaranth (d): Green foxtail (e): Crabgrass (f): Barnyard grass								

Table 26 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(Ic-1)+(B-2) 80 + 250	100: 80: 20	100: 60: 40	90: 0: 90	80: 30: 50	100: 52: 48	100: 30: 70	0
(Ic-1)+(B-3) 80 + 125	100: 95: 5	100: 96: 4	100: 90: 10	40: 30: 10	100: 64: 36	90: 58: 32	0
(Ic-1)+(B-4) 80 + 500	90: 50: 40	100: 60: 40	100: 0: 100	60: 30: 30	60: 40: 20	100: 30: 70	0
(Ic-1)+(B-7) 80 + 125	100: 90: 10	100: 68: 32	40: 20: 20	40: 30: 10	90: 40: 50	100: 30: 70	0
(Ic-1)+(B-11) 80 + 250	80: 50: 30	100: 60: 40	30: 0: 30	100: 72: 28	100: 88: 12	100: 86: 14	0
(Ic-1)+(B-12) 80 + 250	60: 50: 10	100: 60: 40	100: 0: 100	80: 72: 8	100: 76: 24	100: 93: 7	0
(Ic-1)+(B-13) 80 + 250	80: 50: 30	100: 60: 40	100: 0: 100	100: 93: 7	100: 90: 10	100: 90: 10	0
(Ic-1)+(B-18) 80 + 250	100: 60: 40	100: 64: 36	90: 40: 50	100: 79: 21	100: 82: 18	100: 79: 21	0
(Ic-1)+(B-19) 80 + 250	90: 50: 40	100: 60: 40	80: 10: 70	100: 65: 35	100: 82: 18	100: 72: 28	0

(a): Cocklebur (d): Green foxtail

(b): Velvet leaf (e): Crabgrass

(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Table 27 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)						phyto- toxicity
	(a)	(b)	(c)	(d)	(e)	(f)	
	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	(F):(E):(Δ)	corn
(Ic-2)+(B-2) 80 + 250	100: 76: 24	100: 60: 40	100: 0: 100	80: 40: 40	100: 52: 48	100: 30: 70	0
(Ic-2)+(B-3) 80 + 125	100: 90: 10	100: 94: 6	100: 90: 10	40: 0: 40	100: 64: 36	90: 40: 50	0
(Ic-2)+(B-4) 80 + 500	90: 40: 50	100: 60: 40	100: 0: 100	60: 40: 20	60: 40: 20	100: 30: 70	0
(Ic-2)+(B-7) 40 + 125	100: 88: 12	100: 68: 32	40: 20: 20	40: 40: 0	90: 40: 50	100: 30: 70	0
(Ic-2)+(B-11) 80 + 250	80: 40: 40	100: 60: 40	0: 0: 0	100: 76: 24	100: 88: 12	100: 86: 14	0
(Ic-2)+(B-12) 80 + 250	60: 40: 20	100: 60: 40	100: 0: 100	80: 76: 4	100: 76: 24	100: 93: 7	0
(Ic-2)+(B-13) 80 + 250	40: 40: 0	100: 60: 40	100: 0: 100	100: 94: 6	100: 94: 6	100: 93: 7	0
(Ic-2)+(B-18) 80 + 250	90: 52: 38	90: 64: 26	80: 40: 40	100: 82: 18	100: 82: 18	100: 79: 21	0
(Ic-2)+(B-19) 80 + 250	90: 40: 50	90: 60: 30	80: 10: 70	100: 70: 30	100: 82: 18	100: 72: 28	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)
A larger difference (Δ) means a larger synergistic effect
due to the combined use of active ingredients.

Tables 26 and 27 show that all the compositions containing one of Compounds (Ic-1) and (Ic-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing (Ic-1) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18)

and (B-19),

the composition containing Compound (Ic-1) and Compound (B-2) showed a high synergistic effect on slender amaranth, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-) showed a high synergistic effect on in particular, the composition containing Compound (Ic-1) and Compound (B-3) showed a high synergistic effect on barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth, velvet leaf and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-7) showed a high synergistic effect on velvet leaf, crabgrass and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-11) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,

the composition containing Compound (Ic-1) and Compound (B-12) showed a high synergistic effect on slender amaranth, velvet leaf and crabgrass in particular, and

the composition containing Compound (Ic-1) and Compound (B-13), (B-18) or (B-19) showed a high synergistic effect on cocklebur, slender amaranth and velvet leaf in particular.

In the compositions containing (Ic-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ic-2) and Compound (B-2) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ic-2) and Compound (B-3) showed a high synergistic effect on barnyard grass in particular,

the composition containing Compound (Ic-2) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ic-2) and Compound (B-7) showed a high synergistic effect on crabgrass and barnyard grass in particular,

the composition containing Compound (Ic-2) and compound (B-11) showed a high synergistic effect on cocklebur and velvet leaf in particular,

the composition containing Compound (Ic-2) and Compound (B-12) showed a high synergistic effect on slender amaranth and crabgrass in particular,

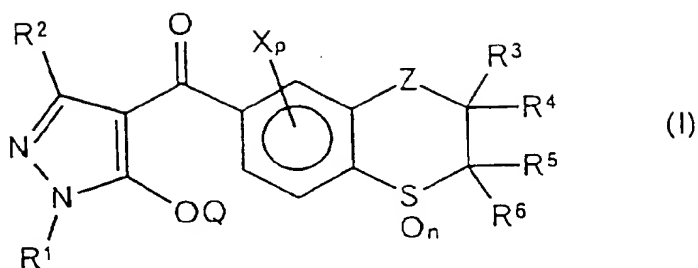
the composition containing Compound (Ic-2) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular, and

the composition containing Compound (Ic-2) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

The herbicide composition of the present invention not only exhibits high herbicidal efficacy but also has a broad herbicidal spectrum on the basis of the synergistic effect of the pyrazole derivative of the general formula (I) and at least one of Compounds (B-1) ~ (B-20) which are active ingredients thereof. Further, the herbicide composition of the present invention exhibits high activity against weeds which are hard to control. Moreover, the herbicide composition of the present invention has high safety for crops such as corn, etc., and is free from damaging (causing phytotoxicity to) crops.

Claims

1. A herbicide composition containing, as active ingredients, a pyrazole derivative of the general formula (I),

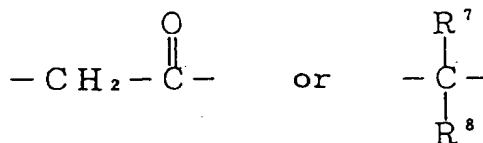
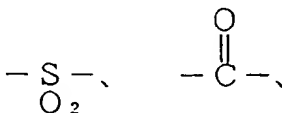


15 {wherein:

20 R¹ is a C₁~C₄ alkyl group, a C₂~C₄ alkenyl group or a C₂~C₄ haloalkenyl group,
 R² is a hydrogen atom, a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group or a C₂~C₄ alkoxyalkyl group,
 X is a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group, a C₂~C₄ alkoxyalkyl group, a halogen atom, a C₁~C₄ alkoxy
 group or a C₁~C₄ haloalkoxy group,
 p 15 an integer of 0, 1 or 2,
 each of R³, R⁴, R⁵ and R⁶ is independently a hydrogen atom, a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group
 or a C₂~C₄ alkoxyalkyl group,
 n is an integer of 0, 1 or 2,
 Q is a hydrogen atom or a group of A-B,

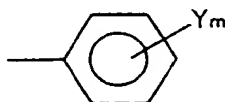
25 [in which

A is a group of



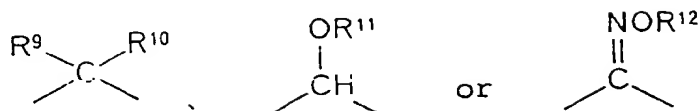
(in which each of R⁷ and R⁸ is independently a hydrogen atom or a C₁~C₄ alkyl group), and

50 B is a C₁~C₁₂ alkyl group, a C₃~C₁₀ cycloalkyl group or a group of



(in which Y is a C₁~C₄ alkyl group, a C₁~C₄ alkoxy group, a C₁~C₄ haloalkyl group, a nitro group or a halo-
 gen atom, and

m is an integer of 0 or 1 ~ 3)), and
Z is



[in which

R⁹ is a hydrogen atom, a C₁~C₄ alkyl group or a C₁~C₄ haloalkyl group,

R¹⁰ is a hydrogen atom, a C₁~C₄ alkyl group, a C₂~C₄ alkenyl group or a C₂~C₄ alkynyl group,

R¹¹ is a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group, a C₃~C₆ cycloalkyl group, a C₃~C₆ alkenylalkyl group, a C₃~C₆ alkynylalkyl group or a C₃~C₆ haloalkenylalkyl group,

R¹² is a C₁~C₄ alkyl group, a C₁~C₄ haloalkyl group, a C₃~C₆ cycloalkyl group, a C₃~C₆ alkenylalkyl group, a C₃~C₆ alkynylalkyl group or a C₃~C₆ haloalkenylalkyl group]],

or a salt thereof; and

at least one herbicide compound selected from the group consisting of

a chloroacetamide-based herbicide,

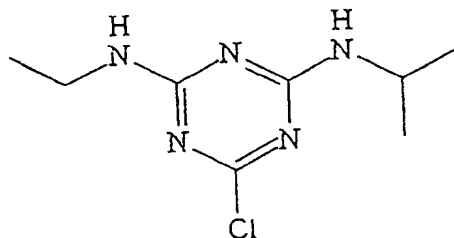
an imidazoline-based herbicide, and

Compound(B-1)

Common name: atrazine

Chemical Name:

6-chloro-N²-ethyl-N⁴-isopropyl-1,3,5-triazine-2,4-diamine

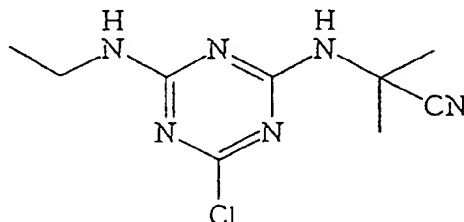


Compound(B-2)

Common name: cyanazine

Chemical Name:

2-(4-chloro-6-ethylamino-1,3,5-triazin-2-ylamino)-2-methylpropionitrile



Compound(B-3)

Common name:metribuzin

Chemical Name:

4-amino-6-tert-butyl-4,5-dihydro-3-methylthio-1,2,4-triazin-5-one

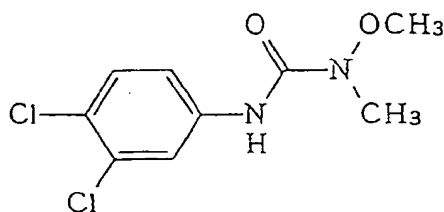


Compound(B-4)

Common name:linuron

Chemical Name:

3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea

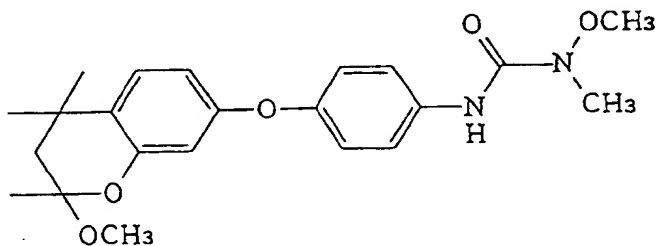


Compound(B-5)

Common name:metbenzurone

Chemical Name:

(±)-1-methoxy-3-[4-(2-methoxy-2,4,4-trimethylchroman-7-yloxy)phenyl]-1-methylurea

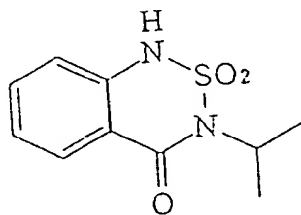


Compound(B-6)

Common name:bentazone

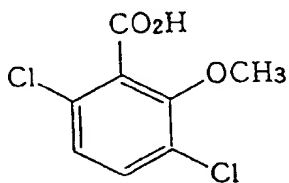
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Chemical Name:
3-isopropyl-1H-2,1,3-benzothiadine-4(3H)-one-2,2-dioxide



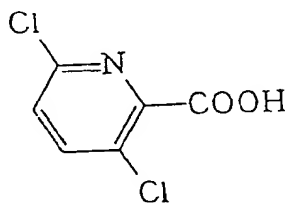
Compound(B-7)

Common name:dicamba
Chemical Name:
3,6-dichloro-2-methoxybenzoic acid



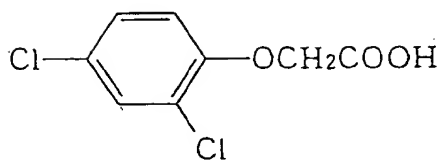
Compound(B-8)

Common name:chlpyralid
Chemical Name:
3,6-dichloropyridine-2-carboxylic acid



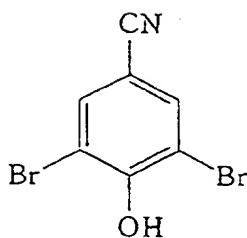
Compound(B-9)

Common name:2,4-D
Chemical Name:
2-(2,4-dichlorophenoxy)acetic acid



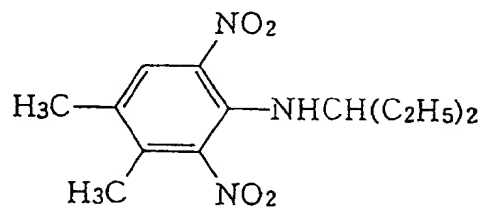
Compound(B-10)

Common name: bromoxynil
Chemical Name:
3,5-dibromo-4-hydroxybenzonitrile



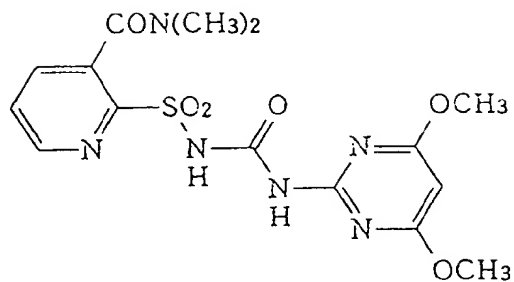
Compound(B-13)

Common name: pendimethalin
Chemical Name:
N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine



Compound(B-14)

Common name: nicosulfuron
Chemical Name:
2-(4,6-dimethoxypyrimidin-2-ylcarbamoylsulfamoyl)-N,N-dimethylnicotineamide

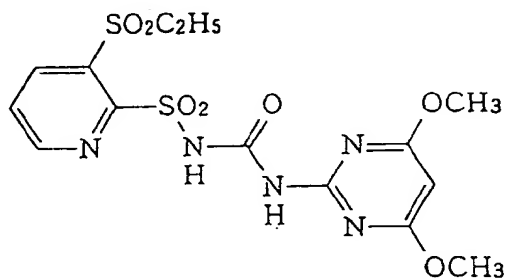


Compound(B-15)

Common name:rimsulfuron

Chemical Name:

1-(4,6-dimethoxypyrimidin-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea

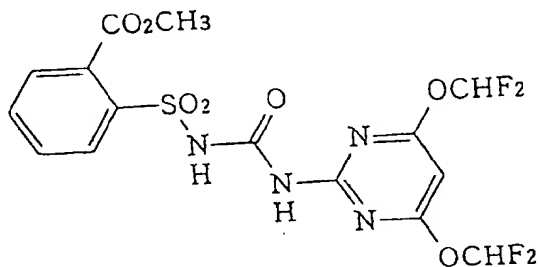


Compound(B-17)

Common name:primisulfuron

Chemical Name:

Methyl 2-[4,6-bis(difluoromethoxy)pyrimidin-2-yl-carbamoylsulfamoyl]benzoic acid



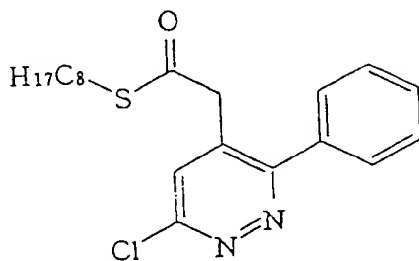
Compound(B-20)

Common name:pyridate

Chemical Name:

EP 0 768 033 A1

6-chloro-3-phenylpyridazin-4-yl-S-octylthiocarbonate



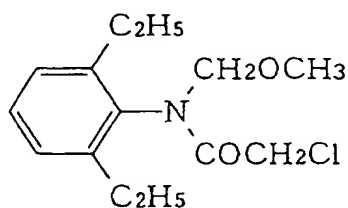
2. The herbicide composition of Claim 1, wherein the chloroacetamide-based herbicide is at least one herbicide compound selected from the group consisting of

Compound(B-11)

Common name:alachlor

Chemical Name:

2-chloro-2',6'-diethyl-N-methoxymethylacetanilide

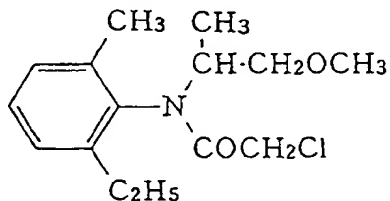


Compound(B-12)

Common name:metolachlor

Chemical Name:

2-chloro-N-(2-ethyl-6-methylphenyl)-N-(2-methoxy-1-methylethyl)acetamide

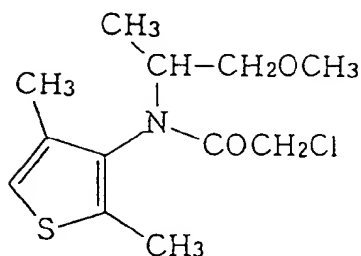


Compound(B-18)

Common name:dimethenamid

Chemical Name:

(1RS,aRS)-2-chloro-N-(2,4-dimethyl-3-thienyl)-N-(2-methoxy-1-methylethyl)acetamide

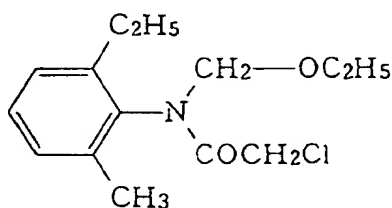


Compound (B-19)

Common name: acetochlor

Chemical Name:

2-chloro-2'-ethyl-6'-methyl-N-ethoxymethylacetanilide



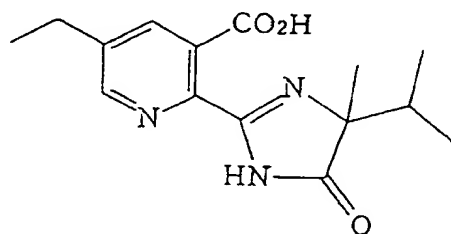
3. The herbicide composition of Claim 1, wherein the imidazoline-based herbicide is at least one herbicide compound selected from the group consisting of

Compound (B-16)

Common name: imazethapyr

Chemical Name:

5-ethyl-2-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-nicotinic acid

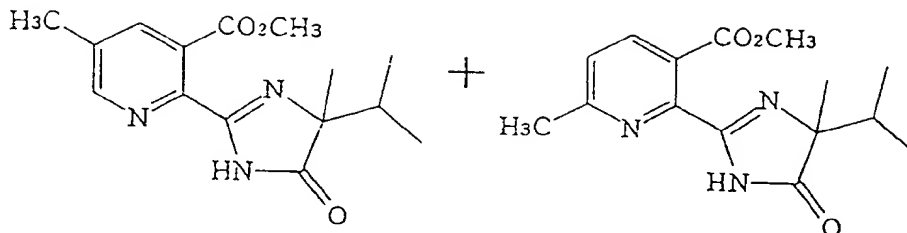


Compound (B-21)

Common name: imazamethabenz-methyl

Chemical Name:

Compound of a Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-m-toluic acid and Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-p-toluic acid.



4. The herbicide composition of Claim 1, wherein in the general formula (I) R^1 is a $C_1\sim C_4$ alkyl group.
5. The herbicide composition of Claim 1, wherein in the general formula (I) R^2 is a hydrogen atom or a $C_1\sim C_4$ alkyl group.
6. The herbicide composition of Claim 1, wherein in the general formula (I) X is a $C_1\sim C_4$ alkyl group or a halogen atom.
7. The herbicide composition of Claim 1, wherein substituent(s) X is/are substituted on the 5-position and/or 8-position of a thiochroman ring.
8. The herbicide composition of Claim 1, wherein in the general formula (I) each of R^3 , R^4 , R^5 and R^6 is independently a hydrogen atom or a $C_1\sim C_4$ alkyl group.
9. The herbicide composition of Claim 1, wherein in the general formula (I) n is 2.
10. The herbicide composition of claim 1, wherein in the general formula (I) Q is a group of -A-B in which B is a group of



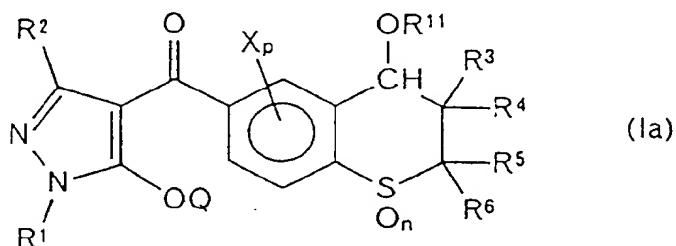
in which Y is a $C_1\sim C_4$ alkyl group, a $C_1\sim C_4$ alkoxy group, a nitro group or a halogen atom.

11. The herbicide composition of any one of Claims 1 to 3, wherein the herbicide composition contains the pyrazole derivative of the general formula (I) and one of Compounds (B-1) ~ (B-20) in the following mixing ratio (weight ratio),

pyrazole derivative(I):compound(B-1:atrazine)=2:1~1:50
 pyrazole derivative(I):compound(B-2:cyanazine)=2:1~1:50
 pyrazole derivative(I):compound(B-3:metribuzin)=3:1~1:25
 pyrazole derivative(I):compound(B-4:linuron)=2:1~1:50
 pyrazole derivative(I):compound(B-5:metbenzuron)=1:2~1:100
 pyrazole derivative(I):compound(B-6:bentazone)=6:1~1:100
 pyrazole derivative(I):compound(B-7:dicamba)=1:1~1:50
 pyrazole derivative(I):compound(B-8:chlpyralid)=4:3~1:12
 pyrazole derivative(I):compound(B-9:2,4-D)=2:1~1:5
 pyrazole derivative(I):compound(B-10:bromoxynil)=1:1~1:50
 pyrazole derivative(I):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(I):compound(B-12:metolachlor)=2:1~1:25
 pyrazole derivative(I):compound(B-13:pendimethalin)=2:1~1:25
 pyrazole derivative(I):compound(B-14:nicosulfuron)=1:3~40:1
 pyrazole derivative(I):compound(B-15:rimsulfuron)=1:3~40:1

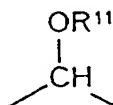
pyrazole derivative(I):compound(B-16:imazethapyr)=1:6~40:1
 pyrazole derivative(I):compound(B-17:primisulfuron)=1:3~ 40:1
 pyrazole derivative(I):compound(B-18:dimethenamid)=2:1~ 1:50
 pyrazole derivative(I):compound(B-19:acetochlor)=2:1~ 1:50
 pyrazole derivative(I):compound(B-20:pyridate)=3:2~ 1:50
 pyrazole derivative(I):compound(B-21:imazamethabenzmethyl)=1:6~40:1

12. The herbicide composition of Claim 1, wherein the pyrazole derivative of the general formula (I) is a pyrazole derivative of the general formula (Ia),

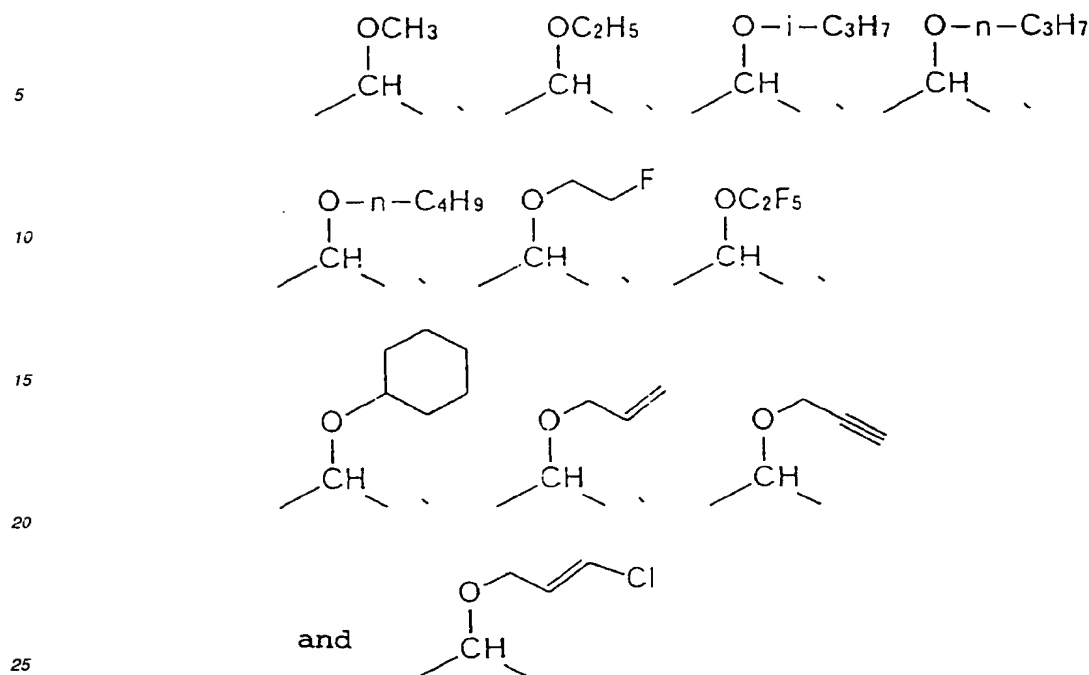


[wherein R¹, R², R³, R⁴, R⁵, R⁶, X, p, n, Q and R¹¹ are as defined in Claim 1]
 or a salt thereof.

13. The herbicide composition of Claim 12, wherein in the general formula (Ia),



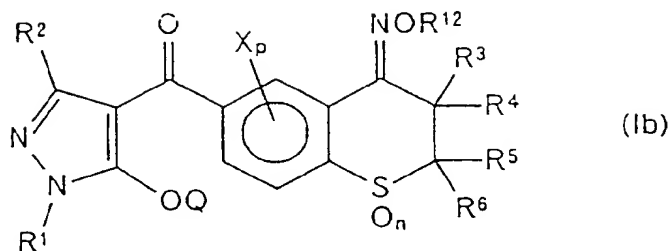
is selected from the group consisting of



14. The herbicide composition of Claim 12, wherein the herbicide composition contains the pyrazole derivative of the general formula (Ia) and one of Compounds (B-1) ~ (B-20) in the following mixing ratio (weight ratio),

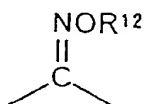
pyrazole derivative(Ia):compound(B-1:atrazine)=1:1~1:50
 pyrazole derivative(Ia):compound(B-2:cyazaflypyr)=2:1~1:25
 pyrazole derivative(Ia):compound(B-3:metribuzin)=3:1~1:12
 pyrazole derivative(Ia):compound(B-4:linuron)=2:1~1:25
 pyrazole derivative(Ia):compound(B-6:bentazone)=1:21~1:100
 pyrazole derivative(Ia):compound(B-7:dicamba)=1:1~1:50
 pyrazole derivative(Ia):compound(B-9:2,4-D)=2:1~1:50
 pyrazole derivative(Ia):compound(B-10:bromoxynil)=1:1~1:50
 pyrazole derivative(Ia):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(Ia):compound(B-12:metolachlor)=2:1~1:25
 pyrazole derivative(Ia):compound(B-13:pendimethalin)=2:1~1:25
 pyrazole derivative(Ia):compound(B-14:nicosulfuron)=1:3~40:1
 pyrazole derivative(Ia):compound(B-15:rimsulfuron)=1:3~40:1
 pyrazole derivative(Ia):compound(B-16:imazethapyr)=1:6~40:1
 pyrazole derivative(Ia):compound(B-17:primisulfuron)=1:3~40:1
 pyrazole derivative(Ia):compound(B-18:dimethenamid)=2:1~1:50
 pyrazole derivative(Ia):compound(B-19:acetochlor)=2:1~1:50
 pyrazole derivative(Ia):compound(B-20:pyridate)=3:2~1:50

15. The herbicide composition of Claim 1, wherein the pyrazole derivative of the general formula (I) is a pyrazole derivative of the general formula (Ib),

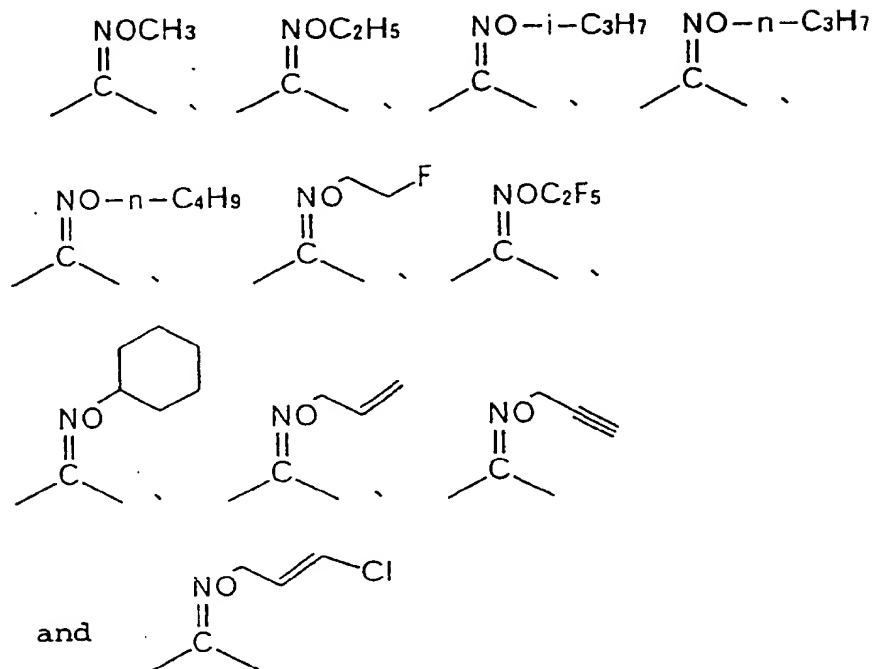


[wherein R¹, R², R³, R⁴, R⁵, R⁶, X, p, n, Q and R¹² are as defined in Claim 1]
or a salt thereof.

16. The herbicide composition of Claim 15, wherein in the general formula (Ib),



is selected from the group consisting of

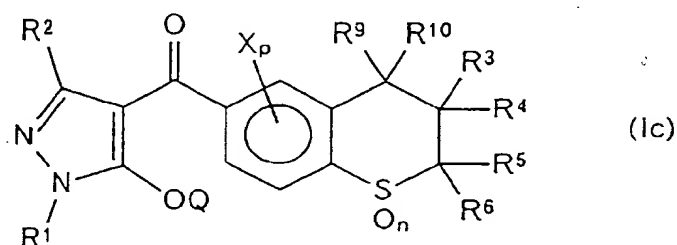


17. The herbicide composition of Claim 15, wherein the herbicide composition contains the pyrazole derivative of the general formula (Ib) and one of Compounds (B-1) ~ (B-19) in the following mixing ratio (weight ratio),

pyrazole derivative(Ib):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(Ib):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(Ib):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(Ib):compound(B-4:linuron)=2:1~1:50

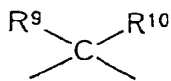
pyrazole derivative(lb):compound(B-5:metbenzurone)=1:2~ 1:100
 pyrazole derivative(lb):compound(B-6:bentazone)=6:1~1:3
 pyrazole derivative(lb):compound(B-7:dicamba)=1:1~1:50
 pyrazole derivative(lb):compound(B-8:chlorpyralid)=4:3~1:12
 pyrazole derivative(lb):compound(B-9:2,4-D)=2:1~1:50
 pyrazole derivative(lb):compound(B-10:bromoxynil)=1:1~1:50
 pyrazole derivative(lb):compound(B-11:alachlor)=2:1~1:25
 pyrazole derivative(lb):compound(B-12:metolachlor)=2:1~ 1:25
 pyrazole derivative(lb):compound(B-13:pendimethalin)=2:1~ 1:25
 pyrazole derivative(lb):compound(B-14:nicosulfuron)=1:3~ 40:1
 pyrazole derivative(lb):compound(B-15:rimsulfuron)=1:3~ 40:1
 pyrazole derivative(lb):compound(B-16:imazethapyr)=1:6~ 40:1
 pyrazole derivative(lb):compound(B-17:primisulfuron)=1:3~ 40:1
 pyrazole derivative(lb):compound(B-18:dimethenamid)=2:1~ 1:50
 pyrazole derivative(lb):compound(B-19:acetochlor)=2:1~1:50

18. The herbicide composition of Claim 1, wherein the pyrazole derivative of the general formula (I) is a pyrazole derivative of the general formula (Ic),

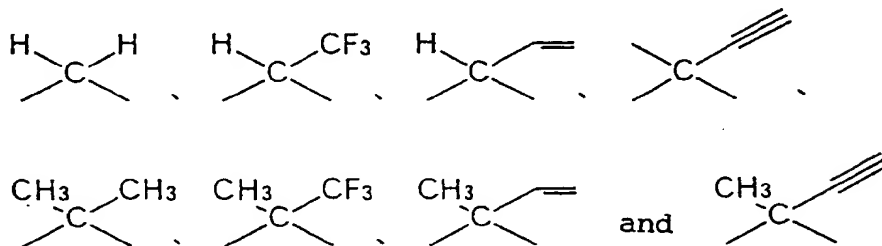


[wherein R¹, R², R³, R⁴, R⁵, R⁶, X, p, n, Q, R⁹ and R¹⁰ are as defined in Claim 1]
 or a salt thereof.

19. The herbicide composition of Claim 18, wherein in the general formula (Ic),



is selected from the group consisting of



20. The herbicide composition of Claim 18, wherein the herbicide composition contains the pyrazole derivative of the general formula (Ic) and one of Compounds (B-1) ~ (B-4) and (B-6) ~ (B-19) in the following mixing ratio (weight ratio),

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pyrazole derivative(lc):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(lc):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(lc):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(lc):compound(B-4:linuron)=2:1~1:50
pyrazole derivative(lc):compound(B-6:bentazone)=1:21~1:100
pyrazole derivative(lc):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(lc):compound(B-8:chlomepyralid)=4:3~1:12
pyrazole derivative(lc):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(lc):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(lc):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(lc):compound(B-12:metolachlor)=2:1~1:25
pyrazole derivative(lc):compound(B-13:pendimethalin)=2:1~1:25
pyrazole derivative(lc):compound(B-14:nicosulfuron)=1:3~40:1
pyrazole derivative(lc):compound(B-15:rimsulfuron)=1:3~40:1
pyrazole derivative(lc):compound(B-16:imazethapyr)=1:6~40:1
pyrazole derivative(lc):compound(B-17:primisulfuron)=1:3~40:1
pyrazole derivative(lc):compound(B-18:dimethenamid)=2:1~1:50
pyrazole derivative(lc):compound(B-19:acetochlor)=2:1~1:50

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP95/01280

A. CLASSIFICATION OF SUBJECT MATTER Int. Cl. ⁶ A01N43/50, A01N43/70, A01N43/707, A01N47/30, A01N43/88, A01N37/40, A01N43/40, A01N37/38, A01N37/34, A01N33/18, A01N47/36, A01N43/58, A01N37/36, A01N43/10 According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) Int. Cl. ⁶ A01N43/50, A01N43/70, A01N43/707, A01N47/30, A01N43/88, A01N37/40, A01N43/40, A01N37/38, A01N37/34, A01N33/18, A01N47/36, A01N43/58, A01N37/36, A01N43/10 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) CAS ONLINE		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO, 94/1431, A1 (Idemitsu Kosan Co., Ltd.), January 20, 1994 (20. 01. 94) & JP, 6-503159, A & CN, 1084170, A	1 - 20
A	WO, 93/18031, A1 (Idemitsu Kosan Co., Ltd.), September 16, 1993 (16. 09. 93) & JP, 5-515530, A & EP, 629623, A1	1 - 20
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.		
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Date of the actual completion of the international search July 18, 1995 (18. 07. 95)		Date of mailing of the international search report August 8, 1995 (08. 08. 95)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.		Authorized officer Telephone No.

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